A MODEL FOR TURBULENT DIFFUSION
IN THE ATMOSPHERE

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A MODEL FOR TURBULENT DIFFUSION

IN THE ATMOSPHERE

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Dedicated to my wife Jaya

whose selfless sacrifices have enchanced the value of this work, to my daughter Seira who had been a continued source of pleasure and to our parents whose encouragements were the beginning of it all.

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NOMENCLATURE

a	Radius of earth
C_p	Specific heat at constant pressure
D	Differential operator
F	Fourier transform
$f(r)$	Longitudinal correlation function
f_y, f_z	The coriolis coordinate $2\Omega \sin\phi$ and $2\Omega \cos\phi$
$g(r)$	Transverse correlation function
g	Acceleration due to gravity
k	Heat transfer parameter
K_x, K_y and K_z	Eddy diffusivity in the three directions
L_x, L_y and L_z	Length scales in the three coordinate directions
$p(\vec{r}_,, \vec{r}_j$	Probability of finding a pair of particles at $\vec{r}_i$ and $\vec{r}_j$
$p(\vec{r})$	Probability of finding pairs of particles at separation $\vec{r}$
$\bar{P}$, P_G	Mean values of pressure at the given height
P'	The perturbed pressure over the mean value
R_{zx}, R_{zy}	Correlation coefficient for a given separation distance in the ZX and ZY planes respectively
R	Gas constant air
$\vec{r}$	Vector separation distance
r_α, r_β and r_γ	The components of separation distance in three directions
$\vec{S}$	Vector separation distance
S_x, S_y	Vertical wind shear $\left(\frac{\partial U}{\partial z}, \frac{\partial V}{\partial z}\right)$

T	Temperature, degrees absolute
$\bar{U}$, u_G	The mean value of X component of velocity
u'	The perturbation velocity in X direction
$\bar{V}$, v_G	The mean value of Y component of velocity
v'	The perturbation velocity in the Y direction
w'	The vertical component of velocity
X, Y and Z	The coordinates in the three directions
α	Specific volume
Γ	The bouyancy term ($dT/dz + g/C_p)/\bar{T}$
ϵ	Turbulent energy dissipation rate
σ_x, σ_y, σ_z	The standard deviations for the particle coordinate in the three directions
σ_u, σ_v, σ_w	The standard deviations for turbulent wind fluctuations
ξ	Time increments
ϕ	Latitude in degrees
ρ	Density
$\bar{\rho}_u$, $\bar{\rho}_v$, $\bar{\rho}_w$	Mean value for the velocity correlation function for the entire cloud
t	Time
Ω	Earth's angular velocity
ω	The proportionality constant for viscous dissipation
λ	Taylor microscale
λ_1	Constant of proportionality for σ_w

Subscripts

i, j	Identification for distinct particles inside the cloud

α, β, γ	Identification for distinct particles inside the cloud
o	Mean value for the entire cloud

Superscripts

$'$	Differentiation with respect to time/turbulent perturbations
—	Mean quantities

Other symbols

$< >$	Mean over the entire cloud

SUMMARY

There have been various methods suggested for the evaluation of air pollution statistics. The K-theory of diffusion has been very widely used for tropospheric air pollution predictions. In many instances the K-theory is able to predict the parameters accurately. This theory is successful mainly in a flow situation where there is one length scale which describes the flow. In the case of the atmosphere there is a spectrum of length scales which affects the flow situation and the use of K-theory cannot be applied with any real justification. The similarity theories have given estimates in those regions where certain similarity parameters could be found, especially inside the atmospheric boundary layers.

In the analysis which is detailed in this thesis another approach is taken which can be used in the atmospheric diffusion studies without being concerned about the limitations of K-theory or similarity approaches. Starting with the equations of motion a set of equations are derived for the variances of the cloud coordinates which are functions of time. This set of equations can be used for different atmospheric stabilities and also in the presence of wind shear. This set gives values for the cross correlations between the cloud coordinates. The various results known earlier are verified by this new scheme.

The diffusion computer program was combined with the Global Reference Atmospheric Model (GRAM) program and has the capability of predicting the diffusion at any latitude longitude point and at heights

varying from surface to 60km and for various times of the year. Many of the assumptions used in the derivation of the equations are typical of stratospheric conditions and are expected to predict the situations at those heights very well.

At lower layers of the atmosphere the diffusion equations were solved and were compared against those obtained by Pasquill-Gifford plots. The agreement appears to be remarkable considering the fact that the Pasquill-Gifford curves themselves are valid only within a range determined by a multiple of two. The various stability effects are reproduced with good accuracy. However, the horizontal dispersion values tend to follow the Taylor results rather than the extrapolation of the Pasquill-Gifford plots.

At stratospheric heights the program was tested against those data compiled by Bauer and the agreement was remarkable. The stability in this region does not undergo any appreciable change and only one stability was tested against the present model.

The capability of the GRAM-Diffusion model to predict long range diffusion calculations were tested for stratospheric heights and for different times of the year. Sample results are shown in Chapter VI.

CHAPTER I

INTRODUCTION

1.1 Review

Study of diffusion and distribution of harmful pollutant materials injected into the atmosphere is one of the major concerns of the present investigations in the field of air pollution. There are many factors which eventually control the concentration of pollutant material at a given location. Atmospheric turbulence is the major driving factor in diffusion. In the atmosphere, diffusion leads to both beneficial and hazardous results. On one hand it can reduce the concentration of contaminants and on the other hand it can diffuse the pollutants to regions where it was absent before. Turbulence and hence diffusion are random at small scales and nothing can be said conclusively about the behavior of a particle from one instant to the other. But on statistical averaging, some properties emerge predictable, to some extent. Theoretical analysis of diffusion has developed mainly on three different approaches. The first and most widely used until recent years was the gradient transport approach. The similarity approach is very recent in the field and requires some more efforts before it could be of wide spread use. The recent and most powerful approach is the statistical approach.

Experimental investigations regarding diffusion are numerous. The atmospheric turbulent diffusion measurements suffer from a main drawback, which often hinders the investigators from verifying any particular theory. It is the unrepeatability of the experiments. The atmospheric conditions cannot be modeled exactly inside a laboratory and one has to rely on the unsteady atmosphere for experiments. Only averaged values of the measurements are of any interest. This fact alone is sufficient to point out that a statistical averaging procedure is more appropriate than any other approach for theoretical investigations of diffusion.

The gradient transport approach was initiated by Adolph Fick, a German physiologist through his paper "Über Diffusion" in 1885. Later on many modifications were made to this basic idea. This theory relates the rate of change of concentration of a mass of passive cloud to the gradient of concentration. This, when mathematically expressed, takes the following form:

$$\frac{d\bar{q}}{dt} = \frac{\partial}{\partial x}\left(K_x \frac{\partial \bar{q}}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial \bar{q}}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_z \frac{\partial \bar{q}}{\partial z}\right)$$

where $\bar{q}$ is the concentration or any conservative property per unit mass and K_x, K_y and K_z are constants or spatially varying, eddy diffusivity coordinates. This particular mathematical form was given by Richardson (1926). Another name which is given to this approach is K-theory. In the special case when the K values are constant a solution in terms of Gaussian functions can be obtained for the governing diffusion equations. The Gaussian nature of concentration distribution is one of the major assumptions employed in many different schemes for the solutions of diffusion problems. The assumption of a constant value of K is inconsistant with the actual observations. Many external factors influence the value of K and modeling of K is very difficult. The work done in this area is enormous and a review of this can be found in Sutton (1953). Priestley (1959) points out that the basic assumption of K-theory is not based on strong experimental evidence. The only justification for its use is that in many cases the results obtained by K-theory agree with actual observations. Recent efforts on diffusion prediction using K-theory are by Runge and Sardei (1975), Ragland and Dennis (1975), Nunge (1974), Lamb, Chen and Seinfeld (1975).

The similarity approach is an important tool in the lower layers of the atmosphere where, to some extent, the scales of motion are specified by the height above the ground. But in this layer turbulence is not homogeneous and a mathematical model for turbulence is extremely difficult. Monin (1959) and Gifford (1962) have modeled diffusion in this lower layer for neutral as well as stratified flow. Since the similarity parameters involved are valid only in the lower layers an alternate modeling becomes essential for higher altitudes.

The recent trend in the studies of diffusion is to follow the statistical theories. This trend was started initially by Taylor (1921). This approach is considerably different from that of the K-theory. Instead of looking at the momentum flux in the Eulerian sense, this method approaches the problem by studying the individual fluid particle to determine its statistical properties. One of the areas the early investigators looked into was the Brownian motion. But the analogy between Brownian motion and atmospheric turbulence was not correct. Brownian motion is the result of completely random phenomenon whereas atmospheric motion consists of highly correlated fluctuations. The study of correlated motions is very involved and they were studied in detail by Goldstein (1951), Lin (1960), Kraichnan (1959), Brier (1950) and many others. One of the major properties which was invariably sought after but never found absolutely was the homogenity of turbulence. This was needed because of the mathematical simplification in the equations governing diffusion process. The assumption of homogenity is approximately valid while treating the inertial subrange of turbulence In the atmosphere there is evidence as shown by MacCready (1962) that the inertial subrange has a wider spectrum of scales of motion than is usually assumed. Moreover, the results of LO-LOCAT experiments as compiled by Atnip (1969) show that even at the low altitudes of 250 feet and 750 feet the homogenity and isotropy of turbulence are remarkable. It has also been shown that the probability distribution for the velocity perturbations are nearly Gaussian. This is a result which is very useful in the mathematical development of the diffusion equations in this thesis. In many cases the results obtained with the assumption of homogenity has been found to be of great practical use. In many instances the ingenuity of investigators has given rise to mathematical formulae for diffusion even though the background theory may not be sound. An example of this is provided by Sutton's (1953) method for atmospheric diffusion which is very widely used for diffusion predictions.

In studying the diffusion of gases, two types of analysis are conducted within the scope of the statistical analysis. One studies the motion of a single particle relative to a fixed axis and the other studies the relative dispersion of the constituent particles to each other. The statistics of the above two kinds of motion are different. In the second case the effect of large scale motions are not felt during the initial period of diffusion when the cluster of particles forming the cloud are of small size. Most of the experimental data recorded are averaged for a number of individual observations and tend to fall under the first type of analysis. The second kind of analysis is more pertinent to study the diffusion of a given cloud of material. Brier (1950) and Kao (1969) examined the second type of analysis and have retrieved the Fickian results asymptotically.

At altitudes above the planetary boundary layer the turbulence is more homogeneous and isotropic than at the lower layers and hence many of the classical results for isotropic turbulence can be utilized for analysis. The stratosphere which extends from roughly 15 km to 45 km altitude has more dominant large scale motions than the troposphere, which occupies the space between the surface and roughly 15 km altitude. In the meridional plane the global circulation has varying aspects in both troposphere and stratosphere. The tropospheric circulation consists of three cells of motion, viz. the Hadley cell (tropical latitude), the Ferrell cell (mid latitude) and the polar cell. Whereas in the stratosphere there is only one cell and this exchanges air mass between the summer and winter hemispheres. The scales of motion in this region are much larger than in the case of the lower region. Most of the diffusing clouds produced in the stratosphere are transported meridionally or zonally by a broad spectrum of waves. Most of these waves might have originated in the troposphere and have moved into the stratosphere. The transient eddies like cyclones get damped when they penetrate into the stratosphere and become standing waves. Thus the wave system in the stratosphere is more or less steady as compared to that in the troposphere.

The diffusing cloud as it is carried around by the mean circulation also increases in volume. As the volume expands larger and larger eddies move some parts of the cloud relative to the other. At the same time physical mixing of the cloud constituents with the atmosphere also occurs. "Mathematical mixing" is the name given to the process of deformation and expansion without physical mixing. According to Danielson (1975) mathematical mixing does not represent diffusion as such. This is in reality an advection process. But most of the stratospheric data are presented after zonal averaging. In doing this averaging these advection motions assume a role equivalent to turbulent motions of large scales. Hence any theoretical model for stratospheric diffusion should consider the large scale motion also as a turbulent fluctuation in order to compare with the observations.

The diffusion as we look at it is scale dependent. When the diffusing cloud is smaller than the size of the dominant eddies the diffusion depends on the scales of motion comparable to the cloud size. This will be the situation as long as the cloud size is smaller than the largest eddy. Once the cloud size becomes greater than the dominant eddies, physical mixing takes place and diffusion can best be explained in terms of the Fickian theory. For large diffusion times the Fickian theory gives a standard deviation

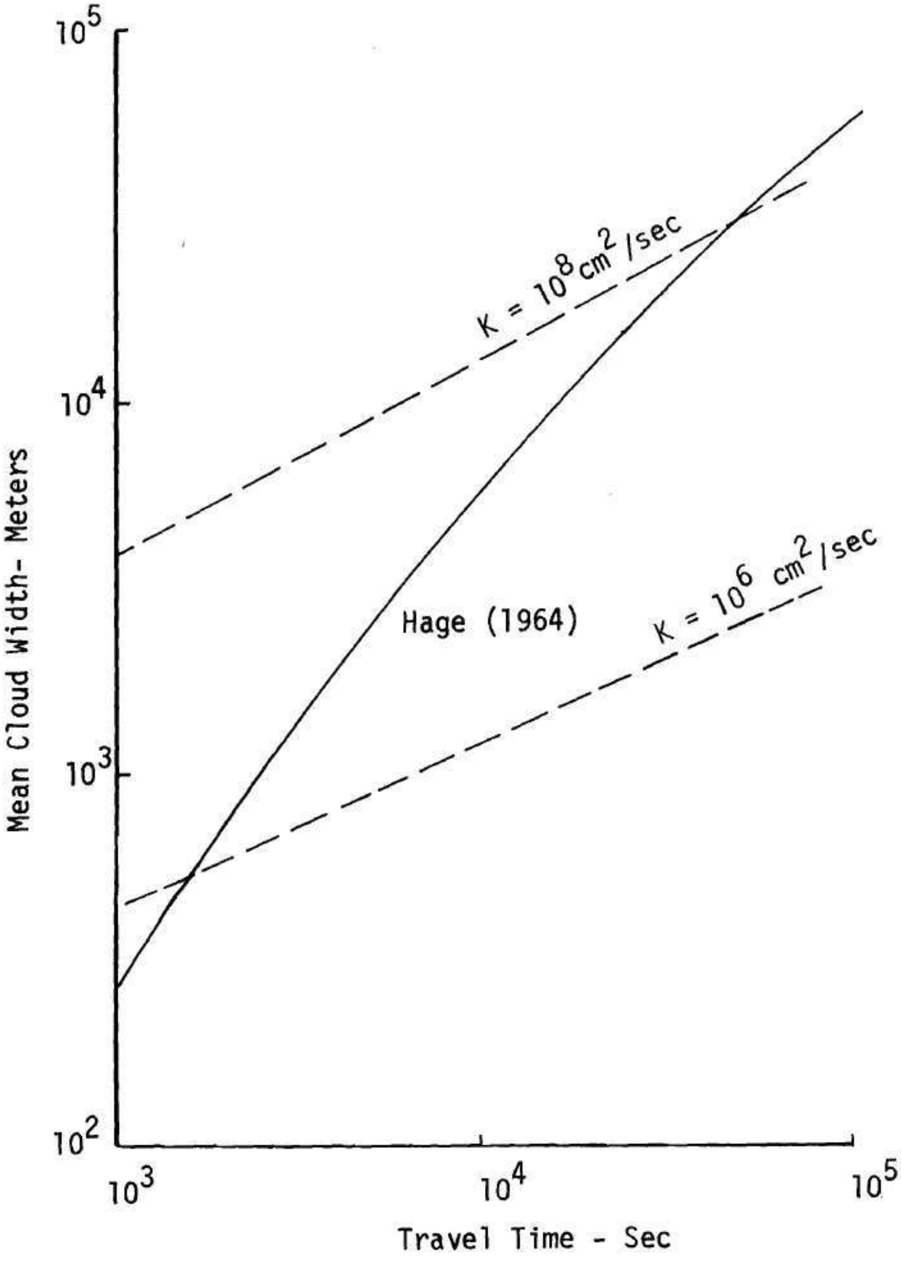


Figure 1. The Scale Dependent Diffusion

$$\sqrt{\overline{< y^2(t) >}} = \sqrt{2Kt}$$

where y(t) is the location of a particle which is averaged over the entire cloud. It can be expected that the statistical analysis also gives the same result. In the present investigation this has been found to be satisfied. When the diffusion is scale dependent the coefficients of diffusion are no longer constant factors. These coefficients change in magnitude as more and more of the spectrum of scales comes into effect. This variation in values in the atmosphere is shown in Figure 1. Atmospheric motion in the stratosphere consists of small scale turbulence as well as large scale waves. As mentioned earlier, the zonal averaging of the diffusion results is equivalent to converting large scale waves to large scale turbulent eddies. This would mean that in the stratosphere the K values would have a large range of values. This violates the basic assumption of proportionality between concentration gradient and rate of change of concentration, used in the Fickian theory.

The statistical approach is more suited for the stratosphere. The stratospheric large scale motions can carry the diffusing cloud to great distances without much diffusion. Thus an analysis with respect to a fixed origin will be unsuited for the situation. The appropriate analysis can be done by means of a relative diffusion approach. Batchelor (1950) has shown a t^2 and t^3 dependence of the variance of cloud size with respect to time. But this is true only when the cloud is in the inertial subrange. This result has been further modified by Walton (1973). These results are obtained on the basis of dimensional reasoning and similarity hypothesis. Hence these results will be found applicable during some time of the diffusion. The t^3 dependence of diffusion process as was obtained on the basis of dimensional reasoning depends more on wind shear effects rather than viscous effects. At small times the wind shear does not influence the cloud size appreciably and a t^3 variation, if it does exist, should be a function of dissipation rate only and would be of secondary importance. The dimensional analysis does not provide any information about the sign of the various terms which build up a series solution. These ambiguities are removed, to some extent, in the present approach to the diffusion problem.

1.2 Present Model

The present formulation of the problem is based on the relative diffusion concept. Instead of studying the field of motion in an Eulerian sense, the dynamics of each particle is studied within the framework of the equations of motion. A similar approach was made by Kao (1962) and (1968). He studied the motion of the particles as consisting of two components. One is the motion of the center of mass and the second is the motion of the particle relative to the center of mass. The viscous terms in the equations of motion are modeled after Rayleigh (1883). The modeling for bouyancy effects is relatively simple. The effect of wind shear is not brought out in detail.

In this present analysis instead of formulating the problem in terms of motion of the center of mass and the relative motion an entirely different approach is made. The basic equations are set up as differences of equations of motion for a pair of particles in the diffusing cloud. This way the study of the mean terms are avoided. The turbulence is then considered as those motions which are superimposed on the geostrophic motions. The viscous terms are modeled in the same way as was done by Kao (1962). The modeling for the bouyancy effects are not assumed to be of a simple form. The heat exchange process is also considered in order to evaluate the bouyancy terms. A correlation between the thermal conductivity and bouyancy was first noted by Monin (1962). He introduced a length scale L* called Bolgiano-Obukov scale. For length scales greater than L* the diffusion was found to depend on the value of heat transfer coefficients. The present model approaches the situation in a different manner. The wind shear effects cannot be neglected while considering long time diffusion. Consideration of wind shear introduces equations for cross correlations between the horizontal and vertical cloud coordinates. The mathematical formulae used for velocity correlations are of the same form as of Batchelor (1959). But they have been modified to suit the different length scales in the vertical and horizontal directions. The concentration of the cloud is modeled in the Gaussian form. This again is normalized with respect to the cloud sizes in the horizontal and vertical directions. The justification for this type of assumption has come from the experimental confirmation by Hay and Pasquill (1957), Cramer, Record and Vaughan (1958) and Barad and Haugen (1959).

The formulation of the diffusion problem in the above method leads to a set of total differential equations, the numerical solution of which does not present any difficulty and Runge-Kutta schemes can be effectively used. The solution depends on the availability of the atmospheric turbulence data. The Global Reference Atmospheric Model developed by Justus, Woodrum, Roper and Smith (1974) was used to obtain most of the turbulence data needed for calculations. As mentioned earlier the viscous terms are modeled in terms of the energy dissipation rate. The availability of these data is very scarce. At present the data obtained by Justus (1966), Justus (1969), Ellsasser (1969) and Kung (1966) and a few others are used. The wide scatter observed in the dissipation rates makes it difficult to choose any particular value.

Experimental studies on dispersion of clouds at various levels in the atmosphere were done by Hage et al (1966), Justus (1966), Heffter (1965), Randerson (1972), Angell (1971), Zimmerman (1966), Greenhow (1959), Blamont and de Jager (1961), Beaudoin et al (1967) and Lilly and Kennedy (1973). The comparison of those results with the present model is discussed towards the end of this thesis. The ability of this diffusion model can be very well tested against the air pollution data at lower altitudes. In this region the presence of different stability classes affects diffusion differently. The vertical and horizontal standard deviations for a diffusing smoke plume have been worked out by Pasquill and those data have been used to test the usefulness of the present formulation. Justus and Mani (1976) have presented these data using an earlier model for the diffusion problem. Since then bouyancy terms have been modeled in terms of an additional heat transfer equation. The final results are discussed in later chapters.

The diffusion model in its present form can be used to study diffusion in the atmosphere from small scales to the very large scales of the stratosphere. The combined computer program of GRAM and Diffusion

Model can handle diffusion at all locations on the globe and for all altitudes up to and including the stratosphere.

CHAPTER II

THE DIFFUSION EQUATIONS

2.1 Equations of Motion

The equations governing the motion of fluid particles are obtained from Newton's relations. In the present formulation we are concerned with the motion of a passive, chemically inert and bouyant cloud element. During the motion of this element, bouyancy and viscosity transforms its kinetic energy to potential and thermal energies. Newton's laws of motion are first written in an inertial coordinate system as shown in Figure 2.

$$\ddot{x}' = F_x' \qquad \ddot{y}' = F_y' \qquad \ddot{z}' = F_z' \tag{2.1}$$

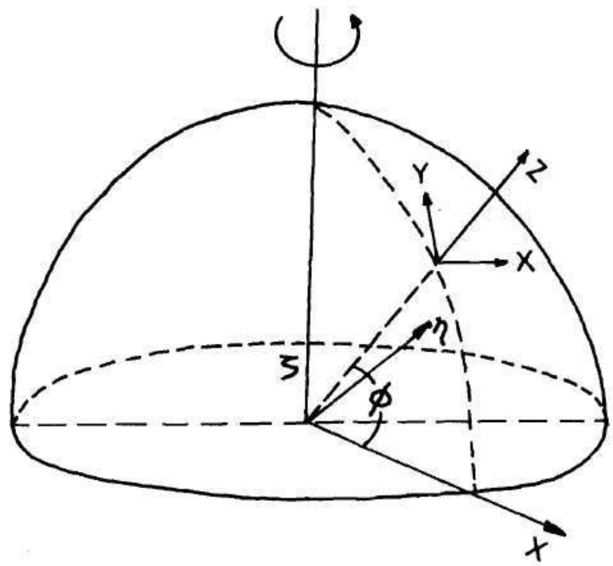
where x, y and z are the coordinates of the particle or element under consideration and F denotes the forces per unit mass in the three coordinate directions. Equations (2.1) are written for an inertial coordinate system. It can be rewritten on a rotating coordinate system ζ, η and χ as follows:

$$\ddot{\chi} = F_x + \chi\Omega^2 + 2\dot{\eta}\Omega \tag{2.2}$$

$$\ddot{\eta} = F_\eta + \eta\Omega^2 - 2\dot{\chi}\Omega \tag{2.3}$$

$$\ddot{\zeta} = F_\zeta \tag{2.4}$$

where Ω is the angular velocity of the coordinate system about the ζ



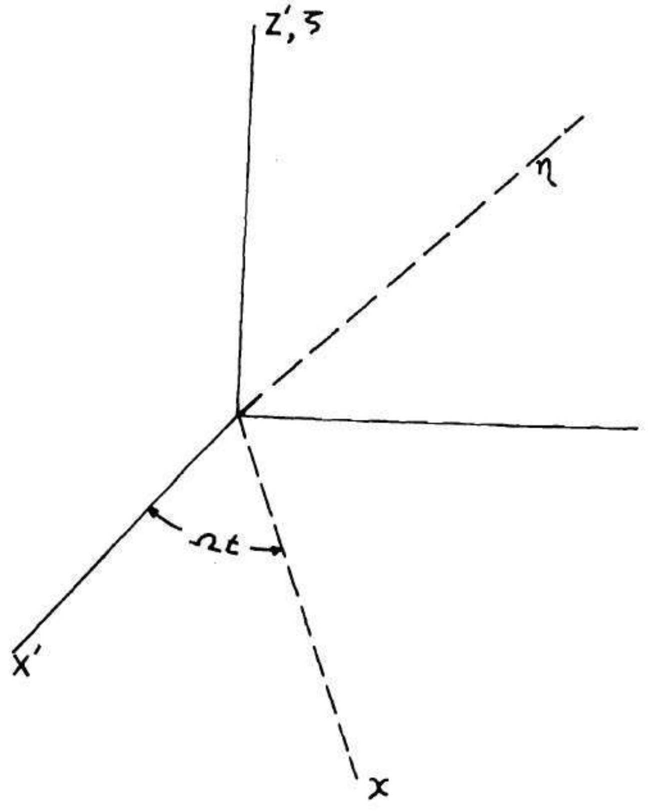


Figure 2. The Coordinate System

axis. The equations (2.2)-(2-4) when written in a coordinate system X, Y, Z as in Figure 2 become equations of motion of particles in a system which is stationary relative to a point on the surface. The components ζ, η and χ and X, Y, and Z are related to each other through the relations

$$\eta = X \tag{2.5}$$

$$\zeta = Z \sin\phi + Y \cos\phi + a \sin\phi \tag{2.6}$$

$$\chi = Z \cos\phi - Y \sin\phi + a \cos\phi \tag{2.7}$$

where a is the radius of the earth. The equations of motion in the new coordinate system become

$$\ddot{X} = 2\Omega(v \sin\phi - w \cos\phi) + F_x \tag{2.8}$$

$$\ddot{Y} = 2\Omega u \sin\phi - a\Omega^2 \sin\phi \cos\phi + F_y \tag{2.9}$$

$$\ddot{Z} = 2\Omega u \cos\phi + a\Omega^2 \cos^2\phi + F_z \tag{2.10}$$

where ϕ is the latitude. The spherical earth is not an equilibrium configuration because a mass at rest on the surface of earth will experience the following accelerations

$$\begin{aligned} \ddot{X} &= 0 \\ \ddot{Y} &= -a\Omega^2 \cos\phi \sin\phi \\ \ddot{Z} &= a\Omega^2 \cos^2\phi - g \end{aligned} \tag{2.11}$$

where g is the acceleration due to gravity. These predict an acceleration towards the equator. The earths surface has been subjected to these forces ever since its formation. The present configuration it has achieved is such that the only acceleration a mass at rest is subjected to is the gravitational acceleration. This deviation from spherical earth model is brought about in the equations of motion by the following substitutions.

$$a\Omega^2 \cos\phi \sin\phi = 0 \tag{2.12}$$

$$a\Omega^2 \cos^2\phi = 0$$

The error introduced due to assumptions given by equation (2.12) are negligible and the resulting equations are universally accepted for atmospheric dynamics calculations. The final form of equations of motion are the following.

$$\ddot{X} = 2\Omega(v \sin\phi - w \cos\phi) + F_x \tag{2.13}$$

$$\ddot{Z} = -2\Omega u \sin\phi + F_y \tag{2.14}$$

$$\ddot{Z} = 2\Omega u \cos\phi - g + F_z \tag{2.15}$$

2.2 Geostrophic Motion

Atmospheric motions consist of turbulence superimposed over mean motions which are global in their extent. In order to study the effect of turbulence the contribution to the equations of motion due to mean motions must be removed. It has been observed that the mean motions are predominantly horizontal relative to the surface of the earth. The external forces acting on a fluid particle are mainly pressure forces and viscous forces. The viscous forces have more effect on the small scale motions. For large scale motions the viscous effects are negligible. But the large scale motion determines the viscous dissipation rates in the small scales of motion. The large scale motions exhibit persistance unlike the small scale motions which are easily damped out. The accelerations of the large scale motions are vanishingly small and hence velocities persist for a long time without change. Introducing these observations mathematically into equations (2.13)-(2.15) certain results can be obtained. The velocity field obtained by the assumption of zero accelerations is known as the geostrophic velocity field.

$$0 = 2\Omega \sin\phi \cdot v_G - \frac{1}{\rho_G} \frac{\partial p_G}{\partial x} \tag{2.16}$$

$$0 = -2\Omega \sin\phi \cdot u_G - \frac{1}{\rho_G} \frac{\partial p_G}{\partial y} \tag{2.17}$$

$$0 = -g - \frac{1}{\rho_G} \frac{\partial pG}{\partial z} \tag{2.18a}$$

$$0 = w_G \tag{2.18b}$$

The term $2\Omega u_G \cos\phi$ was omitted from the last equation (2.18a) in comparison to the other two terms which are more predominant. The geostrophic quantities are functions of the vertical position of the cloud. Atmospheric motion can be assumed to be a superposition of geostrophic motion and turbulent fluctuations. Unlike that in the case of boundary layer turbulence, no assumption is made about the magnitude of the turbulent fluctuations.

2.3 Perturbed Motion

The following decomposition can be applied to all the variables of equations (2.13)-(2.15).

$$\begin{aligned} u &= \bar{U} + u' \\ v &= \bar{V} + v' \\ w &= \bar{W} + w' \end{aligned} \tag{2.19}$$

$$p = \bar{p} + p'$$

$$\alpha = \frac{1}{\rho} = \bar{\alpha} + \alpha'$$

the bar refers to the mean quantities and the prime stands for turbulent fluctuations. The mean quantities are given by the geostrophic equations. Subtracting the mean quantities after substituting equation (2.19) into (2.13)-(2.15) yields the following equations.

$$\frac{d^2X}{dt^2} = f_y v' - f_z w' - \alpha' \frac{\partial \bar{p}}{\partial x} - \bar{\alpha} \frac{\partial p'}{\partial x} - \alpha' \frac{\partial p'}{\partial x} - \omega u' \qquad (2.20)$$

$$\frac{d^2Y}{dt^2} = -f_y u' - \alpha' \frac{\partial \bar{p}}{ay} - \bar{\alpha} \frac{\partial p'}{\partial y} - \alpha' \frac{\partial p'}{\partial y} - \omega v' \qquad (2.21)$$

$$\frac{d^2Z}{dt^2} = f_z u' + f_z \bar{U} - \alpha' \frac{\partial \bar{p}}{\partial z} - \bar{\alpha} \frac{\partial p'}{\partial z} - \omega w' - \alpha' \frac{\partial p'}{\partial z} \qquad (2.22)$$

where

$$f_y = 2\Omega \sin\phi$$

$$f_z = 2\Omega \cos\phi$$

and terms like F_x, F_y and F_z are replaced in the following way

$$F_x = -\frac{1}{\rho} \frac{\partial p}{\partial x} - \omega u' \qquad (2.23)$$

$$F_y = -\frac{1}{\rho} \frac{\partial p}{\partial y} - \omega v' \qquad (2.24)$$

$$F_z = -\frac{1}{\rho} \frac{\partial p}{\partial z} - \omega w' \qquad (2.25)$$

The viscous terms are assumed proportional to the velocity fluctuations as was done by Kao (1962). The nature of the proportionality factor will be investigated later. Equations (2.20)-(2.22) represent the turbulent acceleration of individual particles constituting a given mass of diffusing cloud. Let suffix i denote the i^{th} particle in the given cloud. The equations of motion for the i^{th} particle in the given cloud relative to the mean motion are given by the following relations.

$$\frac{d^2X_i}{dt^2} = -\alpha_i'\frac{\partial\bar{p}}{\partial x} - \bar{\alpha}_i\frac{\partial p_i'}{\partial x} - \alpha_i'\frac{\partial p_i'}{\partial x} - \omega u_i' + f_y v_i' - f_z w_i' \tag{2.26}$$

$$\frac{d^2Y_i}{dt^2} = -\alpha_i'\frac{\partial\bar{p}}{\partial y} - \bar{\alpha}_i\frac{\partial p_i'}{\partial y} - \alpha_i'\frac{\partial p_i'}{\partial y} - \omega v_i' - f_y u_i' \tag{2.27}$$

$$\frac{d^2Z_i}{dt^2} = -\alpha_i'\frac{\partial\bar{p}_i}{\partial z} - \bar{\alpha}_i\frac{\partial p_i'}{\partial z} - \alpha_i'\frac{\partial p_i'}{\partial z} - \omega w_i' + f_z u_i' + f_z\bar{U}_i \tag{2.28}$$

where X_i, Y_i and Z_i are the co-ordinates of the i^{th} particle.

2.4 Relative Dispersion

In order to study the motion of particles relative to each other, the equations of motion have to represent a relation between relative forces and accelerations. This can be obtained by considering the equations of turbulent motion for two distinct particles of the cloud and subtracting one from the other. This differencing eliminates the effects of all mean motions. Consider two points i and j among the cloud particles and designate

$$[\ ]_{ij} = [\ ]_i - [\ ]_j$$

the equations (2.26) through (2.28) can then be rewritten in the following form.

$$\frac{d^2X_{ij}}{dt^2} = -\left[\alpha'\frac{\partial\bar{p}}{\partial x}\right]_{ij} - \left[\bar{\alpha}\,\frac{\partial p'}{\alpha x}\right]_{ij} - \left[\alpha'\frac{\partial p'}{\partial x}\right]_{ij} - \omega u'_{ij}$$

$$+\left[f_y v'\right]_{ij} - \left[f_z w'\right]_{ij} \tag{2.29}$$

$$\frac{d^2Y_{ij}}{dt^2} = -\left[\alpha'\frac{\partial\bar{p}}{\partial y}\right]_{ij} - \left[\bar{\alpha}\,\frac{\partial p'}{\partial y}\right]_{ij} - \left[\alpha'\frac{\partial p'}{\partial y}\right]_{ij} - \omega v'_{ij}$$

$$-\left[f_y u'\right]_{ij} \tag{2.30}$$

$$\frac{d^2Z_{ij}}{dt^2} = -\left[\alpha'\frac{\partial\bar{p}}{\partial z}\right]_{ij} - \left[\bar{\alpha}\,\frac{\partial p'}{\partial z}\right]_{ij} - \left[\alpha'\frac{\partial p'}{\partial z}\right]_{ij} - \omega w_{ij}$$

$$+\left[f_z u'\right]_{ij} + \left[f_z \bar{U}\right]_{ij} \tag{2.31}$$

The equations (2.29)-(2.31) as such cannot be used to evaluate the statistics of the expanding cloud. In order to do that a method has been adopted which has some similarity in procedure as that of Kao (1962) but is new in its philosophy. The equations (2.29)-(2.31) are multiplied by X_{ij}, Y_{ij} and Z_{ij} respectively and are averaged for every individual pair of particles constituting the cloud. This method of averaging has great implications. When an averaging is done for all possible pairs of particles, all possible separation distances are considered and the results does not depend on the correlation functions as a function of separation distance. For cloud sizes comparable to the scales of turbulence these averages become vanishingly small and thereby reduces the complications of closure problems. Mathematically, the above procedure produces the following equations.

$$< X_{ij} \frac{d^2 X_{ij}}{dt^2} > = - < X_{ij} \left[\alpha' \frac{\partial \bar{p}}{\partial x} \right]_{ij} > - < X_{ij} \left[\bar{\alpha} \frac{\partial p'}{\partial x} \right]_{ij} > - < X_{ij} \left[\alpha' \frac{\partial p'}{\partial x} \right]_{ij}$$

$$- < \omega X_{ij} u'_{ij} > + < f_y v'_{ij} X_{ij} > - < f_z \omega'_{ij} X_{ij} > \qquad (2.32)$$

$$< Y_{ij} \frac{d^2 Y_{ij}}{dt} > = - < Y_{ij} \left[\alpha' \frac{\partial \bar{p}}{\partial y} \right]_{ij} > - < Y_{ij} \left[\bar{\alpha} \frac{\partial p'}{\partial y} \right]_{ij} - < Y_{ij} \left[\alpha' \frac{\partial p'}{\partial y} \right]_{ij}$$

$$- < \omega Y_{ij} v'_{ij} > - < Y_{ij} u'_{ij} f_y > \qquad (2.33)$$

$$< Z_{ij} \frac{d^2 Z_{ij}}{dt^2} > = - < Z_{ij} \left[\alpha' \frac{\partial \bar{p}}{\partial z} \right]_{ij} > - < Z_{ij} \left[\bar{\alpha} \frac{\partial p'}{\partial z} \right]_{ij} > - < Z_{ij} \left[\alpha' \frac{\partial p'}{\partial z} \right]_{ij} >$$

$$- < \omega Z_{ij} w'_{ij} > + < f_z u'_{ij} Z_{ij} > + < f_z \bar{U}_{ij} Z_{ij} > \qquad (2.34)$$

where the angle brackets indicate averaging over the cloud. The equations (2.32)-(2.34) cannot be solved as is, and an estimate of the magnitudes of various terms in each of the above equations could possibly bring out the dominant terms. In order to accomplish this result an estimate of ω has to be made initially.

2.5 Estimating ω

In order to estimate ω it is necessary to find out the rate at which kinetic energy is being cascaded from one scale to another scale. The source of this kinetic energy is the mean motion. The turbulent velocities are just carriers of this kinetic energy to the lower scales of motion. Hence, the rate of change of kinetic energy of the velocity fluctuations is equal to the energy dissipation. In order to obtain a relation between the rate of change of kinetic energy and ω the following operations are performed. Equation (2.26) is multiplied by u_i' and is averaged over the entire cloud. It can be noticed that the thermodynamic properties of the atmosphere and the turbulent velocities are very poorly correlated compared to the auto-correlations of the velocities themselves. With this consideration the equation for kinetic energy becomes

$$< \frac{1}{2} \frac{du'^2}{dt^2} > = - < \omega u_i'^2 > + < f_y v_i' u_i' > - < f_z w_i' u_i' > \tag{2.35}$$

Equation (2.27) is similarly multiplied by v_i' and (2.28) by w_i' resulting in the following equations after averaging.

$$< \frac{1}{2} \frac{dv_i'^2}{dt} > = - < \omega v_i'^2 > - f_y < u_i' v_i' > \tag{2.36}$$

$$< \frac{1}{2} \frac{d}{dt} w_i'^2 = - < \omega w_i'^2 > + f_z < u_i' w_i' > + f_z < \bar{U}_i w_i' > \tag{2.37}$$

Equations (2.35)-(2.37) are added together to give the following result.

$$\frac{1}{2}\frac{d}{dt} < u_i'^2 + v_i'^2 + w_i'^2 > = - \omega < u_i'^2 + v_i'^2 + w_i'^2 > + f_z < \bar{U}_i w_i' > \quad (2.38)$$

$\bar{U}_i$ is the mean velocity and in the presence of wind shear, can be represented as the following.

$$\bar{U}_i = U_o + S_x Z_i \quad (2.39)$$

The last term in equation (2.38) becomes

$$f_z < \bar{U}_i w_i' > = f_z < U_o w_i' > + f_z S_x < Z_i w_i' > \quad (2.40)$$

In the case of moderate shear the effect of coriolis accelerations on energy transfer can be neglected in comparison to the energy dissipation. The first term in (2.40) vanishes because of the random nature of w_i'. Thus we obtain the following result.

$$\omega < u_i'^2 + v_i'^2 + w_i'^2 > \quad \gg \quad f_z S_x < z_i w_i' > \quad (2.41)$$

Expressing the rate of energy dissipation per unit mass by ε the following equation can be obtained for ω.

$$< \frac{1}{2}\frac{d}{dr}\left(u_i'^2 + v_i'^2 + w_i'^2\right) > = - \varepsilon = - \omega < u_i'^2 + v_i'^2 + w_i'^2 >$$

$$\omega = \varepsilon / \left(\sigma_u^2 + \sigma_v^2 + \sigma_w^2\right) \quad (2.42)$$

For most atmospheric conditions $\omega \approx 0\ (0.01)/\mathrm{sec}$.

2.6 Equations for Cloud Size

The variances of separation distances can be obtained from equations (2.32)-(2.34). In equations (2.32) and (2.33) the terms which depend on coriolis effects are small compared to the viscous terms on two counts. One because ω is much bigger in magnitude than f_y or f_z and second the correlation between the seperation distance and the velocity separating them is much larger than the correlation between the separation distance and the velocity perpendicular to it. A similar argument holds for equation (2.34) also. The perturbations in the theromodynamic properties are mainly due to the bouyancy effects. The perturbations due to velocity fluctuations are comparatively smaller. Thus in equations (2.32) and (2.33) the correlation between X_{ij} or y_{ij} with the thermodynamic variables can be neglected in comparison to the viscous terms. In equation (2.34) such an assumption is not valid because of the fact that the density perturbations are caused by the bouyancy effects predominantly. Thus there will be a correlation between the vertical components of particle coordinates and the density perturbation. The last term in equation (2.34) requires some more investigation.

$$< f_z \bar{U}_{ij} Z_{ij} > = f_z S_x < Z_{ij} Z_{ij} > = 2 f_z S_x \sigma_z^{\ 2}$$

$$< Z_{ij}^{\ 2} > = < Z'^{\ 2}_{ij} > = < Z'^{2}_{j} > - < 2 Z'_j Z'_i > + < Z'^{2}_{i} >$$

$$= 2 < Z'^{2}_{i} > - 2 < Z'_j Z'_i > \qquad (2.43)$$

where $\qquad Z' = Z - \bar{Z}$

and $\bar{Z}$ is the mean vertical coordinate for the entire cloud. The term $< Z'_j Z'_i >$ vanishes for the cloud on the average. The viscous term in equation (2.39) is rewritten in the following manner.

$$\omega < Z_{ij} w'_{ij} > = \frac{\omega}{2} \frac{d}{dt} < Z_{ij} Z_{ij} > = \omega \frac{d}{dt} \sigma_z^{\ 2} \qquad (2.44)$$

The bouyancy term becomes

$$< Z_{ij} \frac{\alpha'_{ij}}{\bar{\alpha}} g > = \frac{g}{\bar{\alpha}} < Z_{ij} \alpha'_{ij} > \cdot \qquad (2.45)$$

When the stratification of the atmosphere is unstable $\sigma_z^{\ 2}$ and $\frac{d}{dt} \sigma_z^{\ 2}$ are comparable and the coriolis effect is negligible compared to viscous effects. When the stratification is very stable the bouyancy terms are much more predominant than the viscous terms. Again the coriolis effect is negligible as compared to the bouyancy terms. When stratification is neutral the values of wind shear are such that the coriolis term is negligible in comparison to viscous terms, at least up to a time period of $\frac{1}{S_x}$ which is usually high.

The bouyancy affects both the pressure and density of the diffusing cloud. But pressure always equalizes itself with the ambient pressure and the randomness of pressure fluctuations is less dominant than bouyancy induced perturbations. Thus the correlation between the pressure perturbation and the Z_{ij} values vanishes for a large sample.

In equations (2.32) and (2.33) the viscous terms need a closer study. The viscous terms in the equation (2.32) can be rewritten in the following manner.

$$< \omega X_{ij} U'_{ij} > = < \omega X_{ij} U_{ij} > - < \omega X_{ij} \bar{U}_{ij} >$$

$$= < \frac{\omega}{2} \frac{d}{dt} (X_{ij} X_{ij}) > - \omega S_x < X_{ij} Z_{ij} >$$

$$= \omega \frac{d}{dt} \sigma_x^{\ 2} - \omega S_x \sigma_{zx} \qquad (2.46)$$

Similarly the viscous term in equation (2.33) can be written in the following way.

$$\omega < Y_{ij} v'_{ij} > = \omega \frac{d}{dt} \sigma_y^{\ 2} - \omega S_y \sigma_{zy} \qquad (2.47)$$

The left hand side of equation (2.32) can be rewritten in the following form.

$$< X_{ij} \frac{d^2 X_{ij}}{dt^2} > = \frac{1}{2} < \frac{d^2}{dt^2} (X_{ij})^2 > - < \left(\frac{dX_{ij}}{dt}\right)^2 >$$

$$= \frac{d^2}{dt^2} \sigma_x^{\ 2} - < (\bar{U}_{ij} + U'_{ij})^2 > = \frac{d^2}{dt^2} \sigma_x^{\ 2}$$

$$- 2S_x^2 \sigma_x^2 - 2\sigma_u^{\ 2} + 2 < u'_i u'_j > \qquad (2.48)$$

Similar rearrangement can be made for equations (2.33) and (2.34) also, resulting in the following expressions.

$$< Y_{ij} \frac{d^2}{dt^2} Y_{ij} > = \frac{d^2}{dt^2} \sigma_y^{\ 2} - 2S_y^2\sigma_z^2 - 2\sigma_v^2 + 2 < v_i'v_j' > \tag{2.49}$$

$$< Z_{ij} \frac{d^2}{dt^2} Z_{ij} > = \frac{d^2}{dt^2} \sigma_z^2 - 2\sigma_w^2 + 2 < w_i'w_j' > \tag{2.50}$$

By combination of (2.32)-(2.34), (2.44), (2.46), (2.47) and (2.48)-(2.50) the equations for the three variances can be written in the following form.

$$\frac{d^2\sigma_x^2}{dt^2} = 2S_x^2\sigma_z^2 + 2\sigma_u^2 [1 - \bar{\rho}_u] - \omega \frac{d}{dt} \sigma_x^2 + \omega S_x \sigma_{zx} \tag{2.51}$$

$$\frac{d^2\sigma_y^2}{dt^2} = 2S_y^2\sigma_x^2 + 2\sigma_v^2 [1 - \bar{\rho}_v] - \omega \frac{d}{dt} \sigma_y^2 + \omega S_y \sigma_{zy} \tag{2.52}$$

$$\frac{d^2\sigma_z^2}{dt^2} = 2\sigma_w^2 [1 - \bar{\rho}_w] - \omega \frac{d}{dt} \sigma_z^2 - < Z_{ij}\alpha_{ij}' \frac{\partial \bar{p}}{\partial z} > \tag{2.53}$$

where the $\bar{\rho}_{[\ ]}$ stands for $< [\ ]_i'[\ ]_j' >/\sigma_{[\ ]}^2$

Equations (2.51)-(2.53) cannot be solved without further information on σ_{zx}, σ_{zy}, $\bar{\rho}_u$, $\bar{\rho}_v$, $\bar{\rho}_w$ and $< Z_{ij}\alpha_{ij}' \frac{\partial \bar{p}}{\partial z} >$.

2.7 Modeling of $\bar{\rho}_u$, $\bar{\rho}_v$ and $\bar{\rho}_w$

The quantities $\bar{\rho}_u$, $\bar{\rho}_v$, and $\bar{\rho}_w$ are defined in the following way.

$$< [\ ]_i[\ ]_j > = \int_{\vec{S}_i}\int_{\vec{S}_j} [\ ]_i[\ ]_j \ p(\vec{S}_i,\vec{S}_j) \ d\vec{S}_i d\vec{S}_j \tag{2.54}$$

where $\qquad [\ ]_i = [\ ](\vec{S}_i)$

$$[\ ]_j = [\ ](\vec{S}_j)$$

and $$\vec{S}_j = \vec{S}_i + \vec{r}$$

or $$<[\ ]_i[\ ]_j> = \int_{\vec{r}} \overline{[\ ]_i[\ ]_j}\ p(\vec{r})\ d\vec{r} = \int_{\vec{r}} R_{11}(\vec{r})\ p(\vec{r})\ d\vec{r} \qquad (2.55)$$

$p(\vec{S}_i,\vec{S}_j)$ and $p(\vec{r})$ are the probability of finding particles at $\vec{S}_i$ and $\vec{S}_j$ simultaneously and at a separation distance of $\vec{r}$ respectively. The bar denotes average for a given $\vec{r}$ for all values of i in the cloud, which again is denoted as R_{11} or the autocorrelation. Hence the following results are obtained.

$$\bar{\rho}_u = \int_{\vec{r}} R_{11}(\vec{r})\ p(\vec{r})\ d\vec{r} \qquad (2.56)$$

$$\bar{\rho}_v = \int_{\vec{r}} R_{22}(\vec{r})\ p(\vec{r})\ d\vec{r} \qquad (2.57)$$

$$\bar{\rho}_v = \int_{\vec{r}} R_{33}(\vec{r})\ p(\vec{r})\ d\vec{r} \qquad (2.58)$$

The suffix 1, 2 and 3 denote components in the three coordinate directions. The size of the cloud is assumed always to be such that $< u_i'^2 >$ gives the variance of the turbulent field itself. The averaging is different than the ensemble average for a given separation distance. The cloud averaging takes into account a spectrum of separation distances varying between zero and the cloud dimensions. It is often difficult, if not impossible, to define a particular cloud boundary, especially when diffusion has set in. The particles at the edge of the cloud are carried further and further away by turbulence and the edge loses its identity finally. The best way to describe cloud shape in such a situation is to define a cloud particle probability distribution with the boundaries at negative and positive infinities. This way the problem of defining an exact cloud dimension can be reduced to that of defining cloud concentration variances. The averaging will be equal to averaging over the entire physical space but with a weighting function for each coordinate direction. During the initial period of cloud expansion the weighting given by the probability distribution at large distances is very small and $\bar{\rho}_u$, $\bar{\rho}_v$ and $\bar{\rho}_w$ will be very near unity because of the correlation existing between neighboring points. But as time elapses the averaging is done over wider separation distances and the correlation decreases. An exact mathematical experssion for this variation is difficult. Considerable simplification in the mathematical treatment can be brought about if isotropic conditions are assumed, even though isotropy is not realized in practice. But the extent of the inertial subrange in atmospheric turbulence indicates that the best approximation would be that of isotropy.

Consider a pair of particles separated by distance $\vec{r}$. The second order two point velocity correlation function for this situation is given by Batchelor (1959) in the following form.

$$R_{\alpha\beta}(\vec{r}) = \langle u_\alpha u_\beta \rangle = \left[[f(r) - g(r)] \frac{r_\alpha r_\beta}{r^2} + g(r)\ \delta_{\alpha\beta} \right] \overline{u'^2} \qquad (2.59)$$

Where suffix α and β are used to denote the coordinate directions and δ is the Kronecker delta. f and g are scalar functions of separation distances. The significance of functions f and g which are called longitudinal and lateral velocity correlation functions is given in the appendix. r_α and r_β are components of $\vec{r}$ in the directions α and β. The continuity conditions impose a relationship between the longitudinal and lateral correlation functions viz.

$$g(r) = f(r) + \frac{1}{2}\, r\, \frac{df}{dr} \tag{2.60}$$

Substitution of this equation into equation (2.59) gives the following result.

$$R_{\alpha\beta}(\vec{r}) = \overline{u'^2}\left[-\frac{1}{2}\,\frac{f'}{r}\, r_\alpha r_\beta + \left(f + \frac{1}{2} r\, \frac{df}{dr}\right)\delta_{\alpha\beta}\right] \tag{2.61}$$

The prime of f denotes differentiation with respect to r. It is assumed that there are sufficient number of particle pairs at a given separation distance inside the cloud so that equation (2.61) is valid for the average of these particle pairs. This assumption may not be true for large separation distances but this error is reduced by the fact that the weighting at large separation distance is very small. This automatically follows by averaging with a Gaussian distribution function. Equation (2.61) can be used to determine $\bar{\rho}_u$, $\bar{\rho}_v$ and $\bar{\rho}_w$ by averaging it for all the separation distances of the diffusing cloud. In order to do this, it is necessary to have a mathematical expression for concentration distribution of the cloud particles. It is evident from the accumulated experimental evidences that a Gaussian concentration distribution is quite adequate. The probability of finding a particle at position (x,y, z) relative to cloud center of mass is given by

$$p(x,y,z) = \frac{1}{(2\pi)^{3/2}\sigma_x\sigma_y\sigma_z} \exp\left[-\frac{x^2}{2\sigma_x^{\ 2}} - \frac{y^2}{2\sigma_y^{\ 2}} - \frac{z^2}{2\sigma_z^{\ 2}}\right] \qquad (2.62)$$

where σ_x^2, σ_y^2, and σ_z^2 are the variances for cloud sizes and are functions of time for a diffusing cloud. In deriving the mathematical expressions to be used in studying atmospheric turbulence at high altitudes, help is sought from the theory of isotropic turbulence. Even though isotropy cannot be assumed outside of the inertial subrange, there is evidence that the inertial subrange for the atmosphere encloses a wide spectrum of scales. The effect of zonal averaging is to extend the spectrum to the largest scales of motion. Moreover there are observational findings indicating the anisotropy in atmospheric turbulence is well modeled by appropriate changes in the corresponding mathematical expressions for isotropic turbulence. Thus the velocity probability distribution is rewritten in terms of different scales of motion for three coordinate directions. This has the effect of transforming the velocity space into a new velocity space where relations of isotropy could be applied. The relevant velocity correlation in the present analysis is the longitudinal correlation function f(r). f(r) is adequately represented by an exponential variation as below.

$$f(r) = \exp\left[-\frac{\pi}{4}\frac{r^2}{L^2}\right] \qquad (2.63)$$

where L is the length scale for the longitudinal correlation. The value of this quantity is to be determined from measurements. This is in effect the Taylor integral scale of turbulence. By substituting this result into (2.59) the following equation is obtained.

$$R_{\alpha\beta} = \overline{u'^2}\left[\frac{\pi}{4}\frac{1}{L^2}\, r_\alpha r_\beta + \delta_{\alpha\beta}\left(1 - \frac{\pi}{4L^2}r^2\right)\right] \exp\left[-\frac{\pi}{4}\frac{r^2}{L^2}\right] \tag{2.63}$$

$$R_{\alpha\alpha} = \overline{u'^2}\left[\frac{\pi}{4L^2}r_\alpha^2 - \frac{\pi}{4L^2}r^2 + 1\right] \exp\left[-\frac{\pi}{4}\frac{r^2}{L^2}\right] \tag{2.64}$$

The various terms in equation (2.64) are to be modeled appropriately for their analysis. It is assumed that the turbulence is isotropic when normalized with respect to the length scales. Thus the expression for $R_{\alpha\alpha}$ should be a function of r_α/L_α, r_β/L_β and r_γ/L_γ alone, therefore the appropriate forms to choose are the following.

$$r_\alpha^2/L^2 \approx r_\alpha^2/L_\alpha^2 \tag{2.65}$$

$$r^2/L^2 \approx \frac{r_\alpha^2}{L_\alpha^2} + \frac{r_\beta^2}{L_\beta^2} + \frac{r_\gamma^2}{L_\gamma^2} \tag{2.66}$$

Substituting these relations into equation (2.64) we get

$$R_{\alpha\alpha} = \overline{u'^2}\left[1 - \frac{\pi r_\beta^2}{4L_\beta^2} - \frac{\pi r_\gamma^2}{4L_\gamma^2}\right] \exp\left[-\frac{\pi}{4}\left(\frac{r_\alpha^2}{L_\alpha^2} + \frac{r_\beta^2}{L_\beta^2} + \frac{r_\gamma^2}{L_\gamma^2}\right)\right] \tag{2.67}$$

In order to get a mean value for $R_{\alpha\alpha}$ for the entire cloud, probability distriburion for separation $\vec{r} = r_\alpha\vec{\alpha} + r_\beta\vec{\beta} + r_\gamma\vec{\gamma}$ is to be obtained. This can be obtained from the number density for the cloud constituents. The probability of finding a pair of particles at (x_1, y_1, z_1) and (x_2, y_2, z_2) is given by the expression

$$p(\vec{r}_1, \vec{r}_2) = \frac{1}{(2\pi)^3 \sigma_x^2 \sigma_y^2 \sigma_z^2} \exp\left[- \frac{x_1^2 + x_2^2}{2\sigma_x^2} - \frac{y_1^2 + y_2^2}{2\sigma_y^2} - \frac{z_1^2 + z_2^2}{2\sigma_z^2}\right] \quad (2.68)$$

In order to obtain this in terms of separation distance the following substitutions are made

$$x_2 = x_1 + r_x$$

$$y_2 = y_1 + r_y \quad (2.69)$$

$$z_2 = z_1 + r_z$$

The resulting equation is then integrated for all values of x_1, y_1 and z_1 in order to eliminate the dependence on x_1, y_1 and z_1. The resulting equation becomes

$$p(\vec{r}) = \frac{1}{(8\pi)^{3/2} \sigma_x \sigma_y \sigma_z} \exp\left[- \frac{1}{4}\left(\frac{r_x^2}{\sigma_x^2} + \frac{r_y^2}{\sigma_y^2} + \frac{r_z^2}{\sigma_z^2}\right)\right] \quad (2.70)$$

The above equation is very important when studying the cloud behavior in the presence of wind shear. Equation (2.68) is questionable in the presence of wind shear, because of the nonzero values of $< xy >$. But equation (2.70) does not suffer from the same drawback. It can be seen from the following equations that (2.70) is more appropriate than equation (2.68).

$$< x_2 z_2 > = < x_1 z_1 > + < r_x r_z > + < r_z x_1 > + < r_x z_1 > \quad (2.71a)$$

$$< x_2 z_2 > = < x_1 z_1 > \quad (2.71b)$$

$$< r_x Z_1 > = < r_z X_1 > = 0 \tag{2.71c}$$

therefore $$< r_x r_z > = 0 \tag{2.72}$$

The result (2.72) can be obtained directly from equation (2.70). It can be noticed that $< x_2 z_2 >$ will be zero if equation (2.68) is used for evaluation which will not be correct in the presence of wind shear. But an equation in the form (2.70) does not violate any such condition, where wind shear is present because $< r_x r_z >$ vanishes even when wind shear is present. The mean value of $R_{\alpha\alpha}$ for the entire cloud can now be obtained by the following expression.

$$< R_{\alpha\alpha} > = \int_{\vec{r}} R_{\alpha\alpha} p(\vec{r})\, d\vec{r} \tag{2.73}$$

Where the integration is done for all values of $\vec{r}$. By evaluating, the above integral for α = 1,2 and 3. The following results are obtained.

$$\bar{\rho}_u = \frac{C_x C_y C_z}{\sqrt{8}\, \sigma_x \sigma_y \sigma_z} \left[1 - \frac{\pi C_y^2}{4 L_y^2} - \frac{\pi C_z^2}{4 L_z^2} \right] \tag{2.74}$$

$$\bar{\rho}_v = \frac{C_x C_y C_z}{\sqrt{8}\, \sigma_x \sigma_y \sigma_z} \left[1 - \frac{\pi C_x^2}{4 L_x^2} - \frac{\pi C_z^2}{4 L_z^2} \right] \tag{2.75}$$

$$\bar{\rho}_w = \frac{C_x C_y C_z}{\sqrt{8}\, \sigma_x \sigma_y \sigma_z} \left[1 - \frac{\pi C_x^2}{4 L_x^2} - \frac{\pi C_y^2}{4 L_y^2} \right] \tag{2.76}$$

where $$C_{()} = \frac{\sqrt{2}\, \sigma_{()} L_{()}}{\sqrt{\pi \sigma_{()}^2 + L_{()}^2}}$$

It can be seen from equations (2.74)-(2.76) that for large values of L_x, L_y and L_z; $\bar{\rho}_u$, $\bar{\rho}_v$ and $\bar{\rho}_w$ are unity and for large values of σ_x, σ_y and σ_z, ρ_u, ρ_v and ρ_w vanish. This means that during the initial period when the cloud dimension is small compared to the scale lengths $\bar{\rho}_u$, $\bar{\rho}_v$ and $\bar{\rho}_w$ are very close to unity. This is to be expected from the physical nature of the situation. When the cloud dimensions are small most of the constituent particles are separated from each other by a distance so small that the correlation coefficient for that distance is near unity. When averaging is done over the entire cloud which consists of particle pairs of such separation distances, the result is approximately unity. Similarly when the cloud dimensions are large compared to the length scales most of the cloud particles are separated from each other by distances for which the correlation coefficient is vanishingly small. When an averaging for the entire cloud is performed the result is a small number. Hence it can be remarked at this stage that at least qualitatively this model conforms to the physical situation.

2.8 Derivation of Equations for σ_{zx} and σ_{zy}

The determination of σ_{zx} and σ_{zy} are important in the diffusion analysis. When wind shear is present the values of σ_x will be affected by the wind shear more than the value of σ_y if the shear is in the XZ plane. This is because the wind shear moves cloud particles relative to each other. Differential equations for solving these quantities can be obtained in a similar fashion as in the case of σ_x, σ_y and σ_z. In order to obtain an equation for σ_{zx} the X-momentum equation (2.29) is multiplied by Z_{ij} and equation (2.31) is multiplied by X_{ij} and is added along with another term consisting of velocity correlation. This condition is obtained in the following way.

$$\frac{d^2}{dt^2}\sigma_{zx} = < \frac{d^2}{dt^2} Z_{ij}X_{ij} > = < Z_{ij} \frac{d^2}{dt^2} X_{ij} >$$

$$+ < X_{ij} \frac{d^2}{dt^2} Z_{ij} > + 2 < \frac{d}{dt} Z_{ij} \frac{d}{dt} X_{ij} > \qquad (2.77)$$

For unstable stratification the bouyancy terms will be more dominant than either viscous or coriolis terms. Similar conditions persist for very stable stratification also. But for neutral stratification the viscous terms will very much dominate the coriolis effects. Thus, coriolis terms drop out except for those arising from bouyancy and geostrophic balance. Again the correlation between the pressure fluctuations and the cloud coordinates vanishes. Substituting into equation (2.77) from equations (2.29) and (2.31) yields the following result.

$$\frac{d^2}{dt^2} < (Z_{ij}X_{ij}) > = - < Z_{ij}\left[\alpha' \frac{\partial \bar{p}}{\partial x}\right]_{ij} > - < Z_{ij}u'_{ij}\omega > - < X_{ij}\omega w'_{ij} >$$

$$- < X_{ij}\left[\alpha' \frac{\partial \bar{p}}{\partial z}\right]_{ij} > + 2S_x \frac{d}{dt}\sigma_z^2 + 4 < w'_i v'_i >$$

$$- 4 < w'_i u'_j > \qquad (2.78)$$

The fourth term on the right hand side vanishes because of the lack of correlation between the bouyancy induced density perturbation and the X-component of separation distance. The first term on the right hand side gives the bouyancy effects and has to be studied in detail. According to the geostrophic balance equation

$$\bar{\alpha}\,\frac{\partial \bar{p}}{\partial x} = f_y v = f_y[\,v_m + \bar{v} + v'] \tag{2.79}$$

where $$v_m = \langle v \rangle$$

$$\bar{v} = S_y Z$$

thus

$$\langle Z_{ij}\left[\alpha'\frac{\partial \bar{p}}{\partial x}\right]_{ij} \rangle = f_y \langle Z_{ij}\left[\frac{\alpha'}{\bar{\alpha}}\,(v_m + \bar{v} + v')\right]_{ij} \rangle$$

$$= f_y \langle Z_{ij}\left[\frac{\alpha'}{\bar{\alpha}}\right]_{ij} \rangle\, v_m + f_y \langle Z_{ij}\left[\frac{\alpha'}{\bar{\alpha}}\,Z\right]_{ij} \rangle\, S_y$$

$$+ f_y \langle Z_{ij}\left[\frac{\alpha'}{\bar{\alpha}}\,v'\right]_{ij} \rangle \tag{2.80}$$

$$f_y \langle Z_{ij}\left[\frac{\alpha'}{\bar{\alpha}}\right]_{ij} \rangle\, v_m = \langle Z_{ij}\alpha'_{ij} \rangle\, \frac{\partial \bar{p}_m}{\partial x} \tag{2.81}$$

$$\langle Z_{ij}\left[\frac{\alpha'}{\bar{\alpha}}\,Z\right]_{ij} \rangle = 2 \langle Z_i^2\,\frac{\alpha'_i}{\bar{\alpha}} \rangle \tag{2.82}$$

The Z-component of a particle position and the density perturbation are correlated. But for a Gaussian probability distribution for density perturbation the probability of positive and negative values of $\frac{\alpha'}{\bar{\alpha}}$ is equal. Z_i^2 will be always positive and hence in the mean the right hand side of equation (2.82) vanishes.

$$\langle Z_{ij}\left[\frac{\alpha'}{\bar{\alpha}}v'\right]_{ij} \rangle = 2 \langle Z_i\,\frac{\alpha'}{\bar{\alpha}}v' \rangle \tag{2.83}$$

Even if Z_i and α' are correlated, both have negligible correlation with v' and this term also drops out of equation (2.80). A similar analysis can be done with the equations (2.30) and (2.31) giving the equation for σ_{zy}. The final equations to be solved are the following.

$$< \frac{d^2}{dt^2} (Z_{ij}X_{ij}) > = 2S_x\omega\sigma_z^2 - \omega \frac{d}{dt} \sigma_{zx} + 2S_x\frac{d}{dt} \sigma_z^2$$

$$+ 4 < w_i'u_i' > - < Z_{ij}\alpha_{ij}' \frac{\partial \bar{p}}{\partial x} > - 4 < w_i'u_j' > \quad (2.84)$$

$$< \frac{d^2}{dt^2} (Z_{ij}y_{ij}) > = 2S_y\omega\sigma_z^2 - \omega \frac{d}{dt} \sigma_{zy} + 2S_y\frac{d}{dt} \sigma_z^2 + 4 < w_i'v_i' >$$

$$- < Z_{ij}\alpha_{ij}' \frac{\partial \bar{p}}{\partial y} > - 4 < w_i'v_j' > \quad (2.85)$$

Solution of the above equations require information about $< Z_{ij}\alpha_{ij}' >$ and $< w_i'u_i' >$ and $< w_i'v_i' >$.

2.9 Modeling $< Z_{ij}\alpha'_{ij} >$

$< Z_{ij}\alpha_{ij}' >$ is the most important term in equations (2.53), (2.84) and (2.85), when considering the bouyancy effects. The density pertubation due to pressure fluctuations are small in the horizontal plane. The stratification induces density variations which are much higher than random perturbations. The turbulence carries the fluid medium up and down in a random fashion. This vertical motion introduces density fluctuations due to bouyancy which again is random. But the vertical position of a fluid particle will have a relation to its density. Therefore, the correlation between Z_{ij} and α'_{ij} cannot be overlooked. The indicating thermodynamic property for bouyancy effects is the temperature of the cloud element relative to the ambient temperature. The pressure of the cloud element reaches equilibrium with the ambient conditions almost instantly. Let

$$\alpha'_i = \alpha_i - \alpha_{ai} \tag{2.86}$$

where α_i the specific volume of the i^{th} cloud element and α_{ai} is the specific volume of ambient air in contact with the i^{th} element. Which is the same as the mean value of α_i at that height. In the present analysis it is assumed that the cloud material is the same as that of the ambient air. Let the temperature of the cloud particle be denoted by T_i and that of the ambient air by T_{ai}. When the cloud element is thrown around by atmospheric turbulence it undergoes temperature and density changes according to the perfect gas equations. Since the pressure of the cloud particle and the ambient air is the same,

$$\alpha'_i = \frac{T_i - T_{ai}}{T_{ai}} \alpha_{ai} \tag{2.87}$$

Since there exists a temperature difference between the cloud particle and the ambient air a heat transfer process exists which has an effect of relieving the bouyancy influence. The process is extremely complicated. In order to understand the behavior in the mean the following model is used.

$$\frac{dT_i}{dt} = k(T_{ai} - T_i) \qquad (2.88)$$

The proportionality factor k is different from the molecular thermal conductivity and is a method of parameterizing turbulent conductivity and intrainment process plus radiative process which force approach to thermal equilibrium. Some interesting insight can be obtained by observing the Fourier transform of the equation (2.88). In this case α_{ai} and T_{ai} will be fluctuating because the ambient conditions in contact with a fluctuating particle will vary with the location of the particle. The solution of (2.88) in the frequency domain is given by the following equation.

$$\phi_i(\omega) = \phi_{ai}(\omega)/(1 + \omega^2/k^2) \qquad (2.89)$$

where $\phi_i(\omega) = F\{T_i\}$

The F stands for the Fourier transform operator. Equation (2.89) shows that $\phi_i(\omega)$ is negligibly small for $\omega^2/k^2 >> 1$. Thus the contribution to T_i due to high frequencies in T_{ai} is negligible. Only low frequency changes are to be considered while approximately modeling T_i.

The temperature of a particle changes due to two causes. One due to heat transfer and the other due to adiabatic change of pressure due to increase or decrease of height. Consider an adiabatic change for a perfect gas.

$$dQ = C_v dT_A + pd(\frac{1}{p}) = 0 \qquad (2.90)$$

$$C_v dT_A + R\ dT_A = \frac{1}{\rho}\frac{dp}{dz} = -g\ dZ \qquad (2.91)$$

$$C_p dT_A = -gdZ \qquad (2.92)$$

$$dT_A = -g/C_p \; dZ \tag{2.93}$$

When a particle is tossed up and down by turbulence the temperature is changed by both these effects. Hence the total change in temperature can be represented in the following form.

$$dT_i = k(T_{ai} - T_i)\, dt_i - g/C_p dZ_i \tag{2.94}$$

$$d(T_i - T_{ai}) = k(T_{ai} - T_i)\, dt_i - \frac{dT_{ai}}{dZ}\, dZ_i - g/C_p\, dZ_i$$

$$= k(T_{ai} - T_i)\, dt_i - \left(g/C_p + \frac{dT_{ai}}{dZ}\right) dZ_i \tag{2.95}$$

$$g/C_p + \frac{dT_{ai}}{dZ} \approx g/C_p + \frac{d\bar{T}}{dZ} \tag{2.96}$$

The approximation (2.96) is valid because of the fact that the gradient of the fluctuating temperatures is less than the temperature gradient itself. Multiply equation (2.95) by $\alpha_{ai}Z_i/T_{ai}$ and rearrange to obtain the following equation.

$$d\left\{\frac{\left(T_i - T_{ai}\right)\alpha_{ai}Z_i}{T_{ai}}\right\} = k(T_{ai} - T_i)\,\frac{\alpha_{ai}Z_i}{T_{ai}}\, dt_i$$

$$- \left(\frac{dT}{dZ} + g/C_p\right)\frac{\alpha_{ai}Z_i}{T_{ai}}\, dZ_i + (T_i - T_{ai})Z_i d\left(\frac{\alpha_{ai}}{T_{ai}}\right)$$

$$+ (T_i - T_{ai})\,\frac{\alpha_{ai}}{T_{ai}}\, dZ_i \tag{2.97}$$

When the lapse rate $-\frac{dT}{dZ} = \gamma$ is a constant certain simplifications can be made in the equation. The constant lapse rate atmosphere can be represented in the following form.

$$T = T_o - \gamma Z \tag{2.98}$$

Where T_o is a datum temperature. The hydrostatic equation and the perfect gas equation can be combined to give

$$\frac{dp}{dZ} = -\rho g = -pg/(R(T_o - \gamma Z)) \tag{2.99}$$

$$\frac{dp}{p} = -gdZ/(R(T_o - \gamma Z)) \tag{2.100}$$

The above equation when integrated gives

$$\ln(p/p_o) = \frac{g}{R\gamma} \ln\left(\frac{T_o - \gamma Z}{T_o}\right) = \frac{g}{R\gamma} \ln\left(\frac{T}{T_o}\right) \tag{2.101}$$

$$p = \frac{p_o}{T_o}\left[1 - \frac{\gamma Z}{T_o}\right]^{g/R\gamma} \tag{2.102}$$

$$\alpha_{ai}/T_{ai} = R/p_{ai} = \frac{R}{p_o}\left(1 - \frac{\gamma Z_i}{T_o}\right)^{g/R\gamma} \approx \frac{R}{p_o}\left(1 + \frac{gZ_i}{RT_o}\right) \tag{2.103}$$

In equation (2.103) p_o and T_o will be the mean values of pressure and temperature of the cloud. R is the gas constant for air. Even when wind shear is present Z_i can be assumed to have a Gaussian distribution. Z_i and $(T_i - T_{ai})$ are correlated but the correlation between Z_i^2 and $(T_i - T_{ai})$ is vanishingly small because of the randomness of $(T_i - T_{ai})$. The probability of finding a positive $Z_i^2(T_i - T_{ai})$ is the same as the probability of finding a negative $Z_i^2(T_i - T_{ai})$. Z_i^3 also vanishes in the averaging process. The equation (2.97) when averaged over the entire cloud can be written in the following fashion.

$$< \frac{d}{dt} (\alpha' Z_i) > = \frac{d}{dt} < \left[\frac{T_i - T_{ai}}{T_o} Z_i \alpha_o \right] > \tag{2.104}$$

$$\frac{d}{dt} < \frac{T_i - T_{ai}}{T_o} Z_i > = k < \frac{T_{ai} - T_i}{T_o} Z_i > - \frac{\Gamma}{2} \frac{d}{dt} \sigma_z^2$$

$$+ < \frac{T_i - T_{ai}}{T_o} \frac{dZ_i}{dt} > \tag{2.105}$$

Where $$\Gamma = \left(\frac{dT}{dZ} + g/C_p \right) / T_o \tag{2.106}$$

The last term in equation (2.105) is very difficult to evaluate. But certain physical considerations can be utilized, in order to estimate the behavior of this term. The temperature variation is caused by heat transfer and also by bouyancy. The bouyancy component will be very well correlated with the vertical velocity and the heat transfer effect will be negligibly correlated. Let

$$T_i = T_{iB} + T_{iH} \tag{2.107}$$

where suffix B denotes the temperature in the absence of heat transfer and H denotes the perturbation due to heat transfer. According to the previous assumption

$$< T_i \frac{dZ_i}{dt} > \approx < T_{iB} \frac{dZ_i}{dt} > \tag{2.108}$$

Substituting this result into the last term of equation (2.105) the following equation can be obtained.

$$< \frac{T_i - T_{ai}}{T_o} \frac{dZ_i}{dt} > = < \frac{T_{iB} - T_{ai}}{T_o} \frac{dZ_i}{dt} > = \frac{\Gamma}{2} \frac{d\sigma_z^{\ 2}}{dt} \qquad (2.109)$$

This result when substituted into equation (2.105) gives

$$\frac{d}{dt} < \frac{T_i - T_{ai}}{T_o} Z_i > = k < \frac{(T_{ai} - T_i)}{T_o} Z_i > - \Gamma \frac{d}{dt} \sigma_z^2 \qquad (2.110a)$$

or in terms of $\alpha' Z_i$ the following equation can be written.

$$\frac{d}{dt} < \frac{\alpha' Z_i}{\alpha_o} > = k < \frac{\alpha' Z_i}{\alpha_o} > - \Gamma \frac{d}{dt} \sigma_z^2 \qquad (2.110b)$$

Note that the equation (2.110) is exact for two cases of practical interest. When the atmosphere is neutrally stratified

$$T_i = T_{ai} \qquad (2.111)$$

and also

$$\frac{dT}{dZ} = -g/C_p \qquad (2.112)$$

or

$$\Gamma = 0 \qquad (2.113)$$

Thus equation (2.110) is exactly satisfied. A second place where this equation is exact is when heat transfer is zero. In such a case

$$\frac{dT_i}{dz} = - g/C_p \qquad (2.114)$$

$$\frac{dT_{ai}}{dZ} = \frac{dT}{dZ} \qquad (2.115)$$

$$k = 0 \qquad (2.116)$$

Equation (2.110) is again exactly satisfied in this case also.

2.10 Modeling $< w_i' u_i' >$ and $< w_i' v_i' >$

These quantities are nonzero in the presence of Reynolds stress and also when bouyancy is an important mechanism of the turbulence. In the case of isotropic turbulent condition both $< w_i' u_i' >$ and $< w_i' v_i' >$ are zero. The Reynolds stress becomes important in the inertial sublayer of the atmospheric boundary layer. But at all other places, the isotropy condition is more prevalent and the above quantities vanish. The bouyancy induced turbulence will be prevalent in a stationary situation. But in all the conditions of interest in the present investigation there is a mean fluid motion. Hence it is reasonable to assume that on the average for the entire cloud $< w_i' u_i' >$ and $< w_i' v_i' >$ are zero.

2.11 Modeling $< w_i' u_j' >$ and $< w_i' v_j' >$

In the case of isotropic turbulence the above quantities are non-zero for non zero values of separation distance. The correlation function for $w_i' w_j'$ can be approximated in the following way

$$R_{zx} = \sqrt{w'^2 v'^2} \left[\frac{\pi}{4} \frac{r_z}{L_z} \cdot \frac{r_x}{L_x} \right] \exp\left[- \frac{\pi}{4}\left(\frac{r_x^2}{L_x^2} + \frac{r_y^2}{L_y^2} + \frac{r_z^2}{L_z^2} \right) \right] \tag{2.117}$$

the mean value of R_{zx} can be obtained, for the entire cloud, in the following manner.

$$\bar{R}_{zy} = \int R_{zy} P(\vec{r})\, d\vec{r} \tag{2.118}$$

The evaluation of the above integral gives the result

$$< w_i' u_j' > = \bar{R}_{zx} = 0$$

similarly

$$< w_i' v_j' > = \bar{R}_{zy} = 0$$

Thus although w_i' and u_j' or w_i' and v_j' are correlated at specific separations, there is no correlation for $< w_i' u_j' >$ or $< w_i' v_j' >$ when averaged over all separations within the cloud dimensions. The final set of equations to be solved for the cloud dimensions σ_x, σ_y and σ_z and wind shear effects σ_{zx} and σ_{zy} are the following.

$$\frac{d^2}{dt^2}\left[\sigma_x^2\right] = 2S_x^2\sigma_z^2 + 2\sigma_u^2\left[1 - \bar{\rho}_u\right] - \omega\frac{d}{dt}\left[\sigma_x^2\right] + \omega S_x\sigma_{zx} \tag{2.119}$$

$$\frac{d^2}{dt^2}\left[\sigma_y^2\right] = 2S_y^2\sigma_z^2 + 2\sigma_v^2\left[1 - \bar{\rho}_v\right] - \omega\frac{d}{dt}\left[\sigma_y^2\right] + \omega S_y\sigma_{zy} \tag{2.120}$$

$$\frac{d^2}{dt^2}\left[\sigma_z^2\right] = 2\sigma_w^2\left[1 - \bar{\rho}_w\right] - \omega\frac{d}{dt}\left[\sigma_z^2\right] - 2\,\frac{\partial\bar{p}}{\partial z} < \alpha' Z_i > \tag{2.121}$$

$$\frac{d^2}{dt^2}\left[\sigma_{zx}\right] = 2S_x\omega\sigma_z^2 - \omega\frac{d}{dt}\left[\sigma_{zx}\right] + 2S_x\frac{d}{dt}\,\sigma_z^2 - 2\,\frac{\partial\bar{p}}{\partial x} < \alpha' Z_i > \tag{2.122}$$

$$\frac{d^2}{dt^2}\left[\sigma_{zy}\right] = 2S_y\omega\sigma_z^2 - \omega\frac{d}{dt}\left[\sigma_{xy}\right] + 2S_y\frac{d}{dt}\,\sigma_z^2 - 2\,\frac{\partial\bar{p}}{\partial y} < \alpha' Z_i > \tag{2.123}$$

$$\frac{d}{dt} < \frac{\alpha_i' Z_i}{\alpha_o} > = \frac{d}{dt} < \frac{T_i - T_{ai}}{\bar{T}}\, Z_i > = -\,k < \frac{\alpha' Z_i}{\alpha_o} > - \Gamma\,\frac{d}{dt}\,\sigma_z^2 \tag{2.124}$$

Where $\bar{\rho}_u$, $\bar{\rho}_v$ and $\bar{\rho}_w$ are evaluated from (2.74)-(2.76) S_x and S_y are the vertical derivatives of the horizontal components of mean winds, ω is the proportionality constant between viscous forces and perturbation velocity. Γ represents the stability of the atmosphere. It is zero for neutral stability and positive or negative for stable or unstable respectively.

2.12 An Earlier Model

A diffusion model was generated by Justus and Mani (1976). This model was an earlier version of the present model. It did not include the heat transfer equation in the vertical diffusion equation, instead, it used the following model:

$$\alpha'_{ij} = \bar{\alpha}\Gamma(Z_{ij} - \hat{Z}_{ij}) \qquad (2.125)$$

where $\hat{Z}_{ij} = \hat{Z}_i - \hat{Z}_j$ is the difference in equilibrium heights of particle i and j. The term $< \alpha'_{ij} Z_{ij} >$ become

$$< \alpha'_{ij} Z_{ij} > = 2\bar{\alpha}\Gamma\, \sigma_z^2\, (1 - \bar{\rho}_z) \qquad (2.126)$$

where $\bar{\rho}_z = < Z_i \hat{Z}_i > / \sigma_z \sigma_{\hat{z}}$

$\bar{\rho}_z$ is the correlation between particle heights and equilibrium heights. This correlation was modeled in the following way.

$$\bar{\rho}_z = \sigma_z(t/2)/\sigma_z(t) \qquad (2.127)$$

This model could not predict the behavior at very high stabilities. At very high instabilities this model predicted very high diffusion for large down wind distances.

CHAPTER III

THEORETICAL EVALUATION OF THE DIFFUSION EQUATION

3.1 Introduction

The statistical analysis of particle diffusion at large times and also small times were done by Taylor using the properties of correlation functions. The concept of averaging over the cloud was not used; instead, an ensemble average was used for a number of realizations of the same event. Consider X to be the position of a particle at time t, which started from a point where X was zero. This particle was brought to this point with a velocity u' which varied from time to time. Consider a number of repetitions of this event, each time u' being different from before. u' satisfies the condition that a fixed value exists for its variance. The variance of all the values of X so obtained would give the estimate of the size of a diffusing cloud originating from a point source. The time rate of change of this variance gives some interesting results.

$$\frac{d\overline{X^2}}{dt} = 2\,X\,\frac{\overline{dX}}{dt} = 2\,\overline{Xu'} = 2\int_0^t u'(t)\,\overline{u'(t+\xi)}\,d\xi \tag{3.1}$$

If the turbulence is homogeneous and stationary the average value of the product of the velocities can be rewritten in terms of auto correlation functions.

$$\frac{d\overline{X^2}}{dt} = 2\overline{u'^2}\int_0^t R(\xi)\,d\xi \tag{3.2}$$

$$\overline{X^2} = 2\overline{u'^2} \int_o^T \int_o^t R(\xi)\, d\xi dT \tag{3.3}$$

For small values of ξ, $R(\xi)$ is almost unity and the above equation integrates to the following result.

$$\overline{X^2} = 2\overline{u'^2} \cdot \frac{T^2}{2} = \overline{u'^2}T^2 \tag{3.4}$$

For large values of ξ, $R(\xi)$ vanishes and equation (3.3) can be integrated to give the following result.

$$\overline{X^2} = 2\overline{u'^2} \left[\int_o^{t_1} R(\xi)\, d\xi\right] T \tag{3.5}$$

Where t_1 is the value of ξ for which $R(\xi)$ becomes zero. It can be said that for large values of time

$$\overline{X^2}(t) \propto 2\, \sigma_u^2 t \tag{3.6}$$

3.2 Small Time Behavior

A small time analysis cannot be done for a diffusing cloud of negligible size with the diffusion equations. This is because of the fact that many of the assumptions used in the derivation of the diffusion equations are not valid for very small size of the cloud. When the cloud size is small the cloud averaging will not be the same as the ensemble average of the turbulent field. Moreover at zero separation between the various particles, there will be no relative velocity between the particles and there will not be any diffusion. The small time behavior of a cloud of finite size can be found using the diffusion equations in the following way.

At a time close to the initial value the mathematical expressions for $\bar{\rho}_u$, $\bar{\rho}_v$ and $\bar{\rho}_w$ from equation (2.74)-(2.76) assume the following form

$$\bar{\rho}_u = 1 - \frac{\pi\sigma_y^2}{2L_y^2} - \frac{\pi}{2}\frac{\sigma_z^2}{L_z^2} \tag{3.7}$$

$$\bar{\rho}_v = 1 - \frac{\pi}{2}\frac{\sigma_x^2}{L_x^2} - \frac{\pi}{2}\frac{\sigma_z^2}{L_z^2} \tag{3.8}$$

$$\bar{\rho}_w = 1 - \frac{\pi}{2}\frac{\sigma_x^2}{L_x^2} - \frac{\pi}{2}\frac{\sigma_y^2}{L_y^2} \tag{3.9}$$

During the initial periods of diffusion the values of σ_{zx}, σ_{zy} and the bouyancy terms are vanishingly small. Substitution of the above relations into equations (2.119)-(2.121) and a solution in terms of a power series gives the following results.

$$\frac{\sigma_x^2}{L_x^2} = \frac{\sigma_{xo}^2}{L_x^2} + \left\{\left(\frac{2S_x^2L_z^2}{L_x^2} + \frac{\pi\sigma_u^2}{L_x^2}\right)\frac{\sigma_{zo}^2}{L_x^2} + \frac{\pi\sigma_u^2}{L_x^2}\frac{\sigma_{yo}^2}{L_y^2}\right\}\left(t^2 - \frac{\omega t^3}{6}\right) + 0(t^4) \tag{3.10}$$

$$\frac{\sigma_y^2}{L_y^2} = \frac{\sigma_{yo}^2}{L_y^2} + \left\{\left(\frac{2S_y^2L_z^2}{L_y^2} + \frac{\pi\sigma_v^2}{L_y^2}\right)\frac{\sigma_{zo}^2}{L_y^2} + \frac{\pi\sigma_v^2}{L_y^2}\cdot\frac{\sigma_{xo}^2}{L_x^2}\right\}\left(t^2 - \frac{\omega t^3}{6}\right) + 0(t^4) \tag{3.11}$$

Equations (3.10) and (3.11) show that Taylor's results are reproduced for small time behavior. Another important result which can be obtained from the above equations is the ϵt^3 dependence of the diffusion rates, because of the fact that terms like $\omega\sigma_u^2$ and $\omega\sigma_v^2$ in equations (3.10) and (3.11) are replacable by viscous dissipation rate ε, to within a multiplicative constant factor. Batchelor (1951) showed that there is a time period of diffusion where εt^3 variation is observed. The dimensional arguments used did not specify whether the effect was additive or not. It was assumed at that time that it is additive. But the above result shows that the effect is not additive, at least during the initial time. The results shown by equations (3.10) and (3.11) are valid for every stability condition.

3.3 Large Time Behavior

The large time behavior of diffusion equations, in the absence of bouyancy i.e. assuming neutral stability, can be obtained from equation (2.121). Taking into consideration the fact that $\bar{\rho}_u$, $\bar{\rho}_v$ and $\bar{\rho}_w$ each vanish for large time the following result can be obtained. The large time solution to (2.121), neglecting only the constant term σ_{xo} is

$$\sigma_z^2 \approx 2\sigma_w^2 t \tag{3.12}$$

This result is again consistent with the Taylor's result. It has been shown by Gifford (1977), Smith (1965), Saffman (1962) and Corrsin (1953) that the effect of wind shear is to produce a t^3 dependence on the diffusion. In the case of a neutral stratification the solution of equation (2.122) for large time gives the following result.

$$\sigma_{zx} = \frac{2S_x\sigma_w^2 t^2}{\omega} \tag{3.13}$$

Substitution of equation (3.13) into equation (2.119) and integration of the resulting expression gives the result

$$\sigma_x^2 \approx \frac{2}{3} S_x^2 \frac{\sigma_w^2}{\omega} t^3 + \frac{2\sigma_u^2 t}{\omega} \tag{3.14}$$

Where again only the initial constant term has been neglected. The solution (3.14) is a combination of eddy diffusion (i.e. proportional to t) and the empirically observed, shear enhanced t^3 diffusion. The enhanced shear diffusion term formally is the same as was obtained by Smith (1965), except that he derived a 1/6 numerical factor instead of the 2/3 in equation (3.14). Hicks (1971) has shown the existance of a t^3 variation from the numerical simulation approach.

3.4 An Analytical Solution for the Vertical Diffusion, at all Stabilities

The theoretical analysis of the vertical diffusion of a cloud of diffusing particle is difficult in the theoretical studies. Diffusion in the vertical direction depends on the thermal stratification of the cloud. The present formulation has tried to model this important situation in such a way as to distinguish the effects of different classifications of thermal stratification. The solution of equation (2.121) is not very easy. Equation (2.121) and (2.124) can be combined together to give one equation in terms of σ_z. In order to combine (2.121) and (2.124) equation (2.124) is multiplied by exp [kt] to obtain the following result:

$$\frac{d}{dt}\left\{e^{kt} < \frac{\alpha_i' Z_i}{\alpha_o} >\right\} = - I\left[\frac{d}{dt}\,\sigma_z^2\right] e^{kt} \tag{3.15}$$

Equation (2.121) is similarly multiplied by e^{kt} and differentiated once with respect to t, giving the following result.

$$e^{kt}\frac{d^3}{dt^3}\left[\sigma_z^2\right] + ke^{kt}\frac{d^2}{dt^2}\left[\sigma_z^2\right] = 2\sigma_w^2\frac{d}{dt}\left\{e^{kt}\left[1 - \bar{\rho}_w\right]\right\} - \omega\frac{d}{dt}\left\{e^{kt}\frac{d}{dt}\left[\sigma_z^2\right]\right\}$$

$$- 2g\frac{d}{dt}\left\{< \frac{\alpha' Z_i}{\alpha_o} > e^{kt}\right\} \tag{3.16a}$$

Equation (2.14a) can be substituted into (2.14b) to yield,

$$X = \sigma_z^2 \tag{3.16b}$$

$$X''' + (k + \omega)X'' + (\omega k + 2g\Gamma)X' = 2\sigma_w^2\frac{d}{dt}\left[1 - \bar{\rho}_w\right]$$

$$+ 2\sigma_\omega^2 k\left[1 - \bar{\rho}_w\right] \tag{3.17}$$

where the primes indicate differentiation with respect to time. Equation (3.17) can be rewritten in the following form (after integrating once).

$$(D - \beta_1)(D - \beta_2)X = 2\sigma_w^2\left[1 - \bar{\rho}_w\right] + 2\sigma_w^2 k\int_o^t\left[1 - \bar{\rho}_w\right]dt' + I \tag{3.18}$$

Where D is a differential operator and I is a constant of integration. The solution to the differential equation (3.18) can be written in a general form without making any assumptions regarding $\bar{\rho}_w$ thus:

$$\frac{X(\beta_1-\beta_2)}{2\sigma_w^2} = e^{\beta_1 t}\int_o^t e^{-\beta_1 t'}\left[1 - \bar{\rho}_w\right]dt' + ke^{\beta_1 t}\int_o^t e^{-\beta_1 t'}\int_o^{t'}\left[1 - \bar{\rho}_w\right]dtdt'$$

$$- e^{-\beta_2 t}\int_o^t e^{-\beta_2 t'}\left[1 - \bar{\rho}_w\right]dt' - ke^{\beta_2 t}\int_o^t e^{-\beta_2 t'}\int_o^{t}\left[1 - \bar{\rho}_w\right]dtdt'$$

$$+ Ae^{\beta_1 t} + Be^{\beta_2 t} + E \tag{3.19a}$$

where

$$\beta_1 = -(k+\omega) + \sqrt{(k+\omega)^2 - 4(\omega k + 8g\Gamma)} = -(k+\omega)$$

$$+ \sqrt{(k-\omega)^2 - 8g\Gamma} \tag{3.19b}$$

and $$\beta_2 = -(k+\omega) - \sqrt{(k-\omega)^2 - 8g\Gamma} \tag{3.19c}$$

Equation (3.19) can be rewritten in the following form

$$\frac{X(\beta_1 - \beta_2)}{2\sigma_\omega^2} = e^{\beta_1 t}\int_o^t e^{-\beta_1 t'}\left[1 - \bar{\rho}_w\right]dt' - \frac{ke^{\beta_1 t}}{\beta_1}\int_o^t\left[1 - \bar{\rho}_w\right]\left[e^{-\beta_1 t}\right.$$

$$\left. - e^{-\beta_1 t}\right]dt - e^{\beta_2 t}\int_o^t e^{-\beta_2 t'}\left[1 - \bar{\rho}_w\right]dt'$$

$$+ \frac{ke^{\beta_2 t}}{\beta_2}\int_o^t\left[1 - \bar{\rho}_w\right]\left[e^{-\beta_2 t} - e^{-\beta_2 t}\right]dt + Ae^{\beta_1 t}$$

$$+ Be^{\beta_2 t} + E \tag{3.20}$$

or

$$\frac{X(\beta_1-\beta_2)}{2\sigma_\omega^{\ 2}} = \left[1 + \frac{k}{\beta_1}\right] e^{\beta_1 t} \int_o^t e^{-\beta_1 t'} \left[1 - \bar{\rho}_w\right] dt' - \frac{k}{\beta_1} \int_o^t \left[1 - \bar{\rho}_w\right] dt$$

$$- \left[1 + \frac{k}{\beta_2}\right] e^{\beta_2 t} \int_o^t e^{-\beta_2 t} \left[1 - \bar{\rho}_w\right] dt'$$

$$+ \frac{k}{\beta_2} \int_o^t \left[1 - \bar{\rho}_w\right] d\tau + Ae^{\beta_1 t} + Be^{\beta_2 t} + E \tag{3.21}$$

The equation (3.21) can be written in a definite form after evaluating constants A, B and E. The boundary conditions are specified at t equal to zero. The starting value of the diffusing cloud is usually known. The first derivative of the variance is assumed to be zero. This is because of the fact that during the initial period of diffusion it has been observed that the variation is proportional to at least the second power of time. The equation (3.17) requires a condition on the second derivative of X in order to obtain a solution. The initial value of $< \alpha_i' Z_i >$ is zero because at the beginning the particles are in a state equilibrium. Equation (2.121) indicates that to be consistent

$$X''\Big|_{t=0} = 2\sigma_\omega^{\ 2}(1 - \eta) \tag{3.22}$$

where η is the initial value of $\bar{\rho}_w$. When the above considerations are applied to equation (3.21) the following solution results.

$$\frac{X(\beta_1 - \beta_2)}{2\sigma_\omega^{\ 2}} = \left[1 + \frac{k}{\beta_1}\right] e^{\beta_1 t} \int_o^t e^{-\beta_1 t} \left(1 - \bar{\rho}_w\right) dt$$

$$- \frac{k}{\beta_1} \int_o^t \left(1 - \bar{\rho}_w\right) dt$$

$$- \left[1 + \frac{k}{\beta_2}\right] e^{\beta_2 t} \int_o^t e^{-\beta_2 t}\left(1 - \bar{\rho}_w\right) dt$$

$$+ \frac{k}{\beta_2} \int_o^t \left(1 - \bar{\rho}_w\right) dt + \frac{X_o(\beta_1 - \beta_2)}{2\sigma_w^{\ 2}} \qquad (3.23)$$

Equation (3.23) in the present form is not complete because the variation of $\bar{\rho}_w$ with time is not yet known. Until then only the behavior at large time alone can be obtained from equation (3.23). At large values of time $\bar{\rho}_w$ vanishes because the size of the cloud becomes larger than the scale of turbulence. In this situation the following assumption can be made

$$\int_o^t F(t) \left(1 - \bar{\rho}_w\right) dt \approx \int_o^t F(t) dt \qquad (3.24)$$

with this substitution equation (3.23) becomes

$$\frac{X_\infty(\beta_1 - \beta_2)}{2\sigma_w^{\ 2}} = \left(1 + \frac{k}{\beta_1}\right)\left(- \frac{1}{\beta_1}\right)\left[1 - e^{\beta_1 t}\right] - \frac{kt}{\beta_1} + \frac{kt}{\beta_2}$$

$$- \left(1 + \frac{k}{\beta_2}\right)\left(- \frac{1}{\beta_2}\right)\left[1 - e^{\beta_2 t}\right] + \frac{X_o(\beta_1 - \beta_2)}{2\sigma_w^{\ 2}} \qquad (3.25)$$

The variation of X_∞ will depend on the values of β_1 and β_2.

3.4.1 Very Stable Stratification

When the stratification is very stable, Γ is positive. Since ω and k are positive values the values of β_1 and β_2 both will be negative. From equation (3.19b) and (3.19c) it can be seen that

$$-(k+\omega) < \pm\sqrt{(k+\omega)^2 - 4(\omega k + 2g\Gamma)}$$

$$-(k+\omega) \mp \sqrt{(k+\omega)^2 - 4(\omega k + 2g\Gamma)} < 0 \tag{3.26}$$

when t becomes very large the exponential terms in equation (3.25) drop out. The term containing variation with t becomes much larger than the constant terms giving

$$X_\infty \approx \frac{2\sigma_\omega^2 k}{\omega k + 2g\Gamma}\, t \tag{3.27}$$

The linear dependence of the vertical variance of cloud size with time has been observed in experiments also. Another important consequence of equation (3.25) is the behavior when

$$2g\Gamma \ll \omega k$$

In this case the asymptotic behavior of X becomes

$$X_\infty \approx \frac{2\sigma_w^2}{\omega}\, t \quad \text{when } 2g\Gamma \ll \omega k \tag{3.28}$$

Equation (3.28) is the same as equation (3.8). This indicates that as the stability tends to neutral conditions the dependence on the value of k is decreased. In fact 'k' is a parameter which indicates the bouyancy effects on the diffusion. The neutral conditions do not produce any bouyancy effects.

3.4.2 Very Unstable Stratification

When the stratification is unstable the value of Γ will be negative. In addition if

$$|2g\Gamma| > \omega k \tag{3.29}$$

then
$$(k + \omega) < \pm\sqrt{(k + \omega)^2 - 4(\omega k + 2g\Gamma)}$$

or
$$-(k + \omega) \pm\sqrt{(k + \omega)^2 - 4(\omega k + 2g\Gamma)} > 0 \tag{3.30}$$

When t becomes very large the exponential terms become much larger than the linear terms and the constant terms. From equation (3.23) it can be seen that

$$X_\infty \approx \frac{1}{\beta_1}\left(1 + \frac{k}{\beta_1}\right) e^{\beta_1 t} + \frac{1}{\beta_2}\left(1 + \frac{k}{\beta_2}\right) e^{\beta_2 t} \tag{3.31}$$

It is observed in practice that the very unstable condition produces diffusion in the vertical direction which varies exponentially. In this case k plays an important part in the diffusion.

3.4.3 Neutral Stratification

As was observed earlier the variation of σ_z, in the case of neutral stratification and at very large time, should be like that for σ_x and σ_y. The neutral condition is mathematically expressed as

$$\Gamma = 0 \tag{3.32}$$

This indicates that

$$\beta_1 = - (\omega + k) + \sqrt{(k - \omega)^2} = - 2\omega \tag{3.33}$$

$$\beta_2 = - (\omega + k) - (k - \omega) = - 2k \tag{3.34}$$

The values of β_1 and β_2 both are negative and at large values of t the exponential terms drop out. The linear terms will dominate. Hence

$$X_\infty = \frac{2\sigma_w^2}{\omega} t \tag{2.35}$$

This result was obtained earlier for the case of horizontal variance. This result can be obtained by inspection of equation (2.121) and (2.124) also. When the stratification is neutral the solution of equation (2.124) gives zero value for $< \alpha_i' Z_i >$ for all time. With this result equation (2.121) will be the same as equation (2.119) and (2. 120) in the absence of wind shear.

3.5 Representation of $\bar{\rho}_w$

The analytical expression for $\bar{\rho}_w$ involves σ_x, σ_y and σ_z which are functions of time. Hence $\bar{\rho}_w$ cannot be in general expressed in closed form as a function of time alone. Without such an expression the equation for σ_z^2 cannot be evaluated in closed form. An approximation as in the following equation appears to be appropriate.

$$\bar{\rho}_w = \eta \exp [- \alpha t^2] \tag{3.36}$$

where η is the value of $\bar{\rho}_w$ at time zero and α is a parameter which at the most depends on the initial conditions and is independent of time. The exact dependence of α on the initial values is not known precisely but all the computations carried out so far indicates that the equation (3.36) appropriately represents the variation of $\bar{\rho}_w$. The use of equation (3.36) in equation (3.23) provides a solution for σ_z^2 as a function of time in terms of the parameter α, and the behavior of σ_z^2 during various stages of diffusion can be obtained from equation (3.23).

CHAPTER IV

THE DIFFUSION EXPERIMENTS

4.1 Pasquill Gifford Diffusion Estimates

The Pasquill-Gifford diffusion plots are empirical results and are derived from various experiments and also from theoretical estimates. The main success of the method lies in the fact that the stratification of the atmosphere has been grouped into stability classifications based on the value of lapse rate, wind speed and solar insolation. Table 1 below indicates the dependence of the stability classification on various factors. The Pasquill-Gifford plots are shown in Figures 3 and 4. They represent the change in values of σ_x, σ_y and σ_z with down wind distance as the cloud is carried downstream with the wind. There exists a different plot for each stability classification indicating a strong dependence on the value of Γ. Figure 3 shows the variation of σ_y with down wind distance. The values of σ_y for large downwind distances were obtained by extrapolation of the values at low downwind distances. But this trend is not correct and instead a variation of σ_y^2 proportional to the downwind distance was in fact found to be more appropriate. This result was obtained earlier by equation (3.3). It can be noticed that during the initial periods of diffusion the variation of σ_y^2 proportional to the square of the downwind distance is more predominant. This agrees with the Taylor's results for point sources. These plots can be effectively used only for the small downwind distances. Moreover, the above

TABLE 1: The Dependence of
Stability Classification on Various Factors

Pasquill Class	Description	$\Delta T\ ^{\circ}C$/100 meters		
A	Extremely Unstable	≤	to	-1.9
B	Unstable	-1.8	to	-1.7
C	Slightly Unstable	-1.6	to	-1.5
D	Neutral	-1.4	to	-0.5
E	Slightly Stable	-0.4	to	+1.4
F	Stable	1.5	to	3,9

Wind Speed at 10m	Insolation			Night	
	Strong	Moderate	Slight	≥ 4/8	≤ 3/8
2	A	A-B	B	F	F
2-3	A-B	B	C	E	F
3-5	B	B-C	C	D	E
5-6	C	C-D	D	D	D
6	C	D	D	D	D

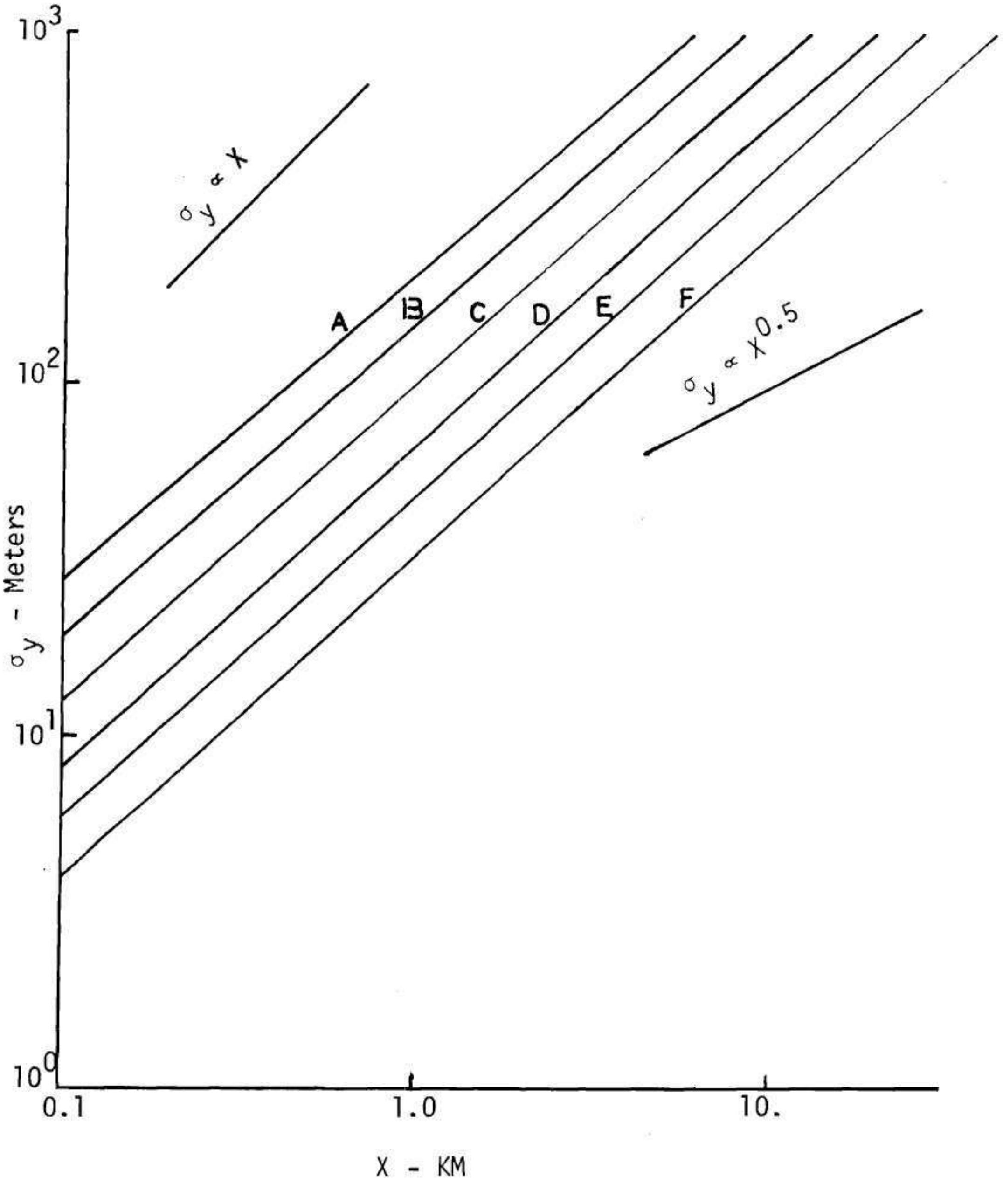


Figure 3. Horizontal Dispersion Standard Deviation

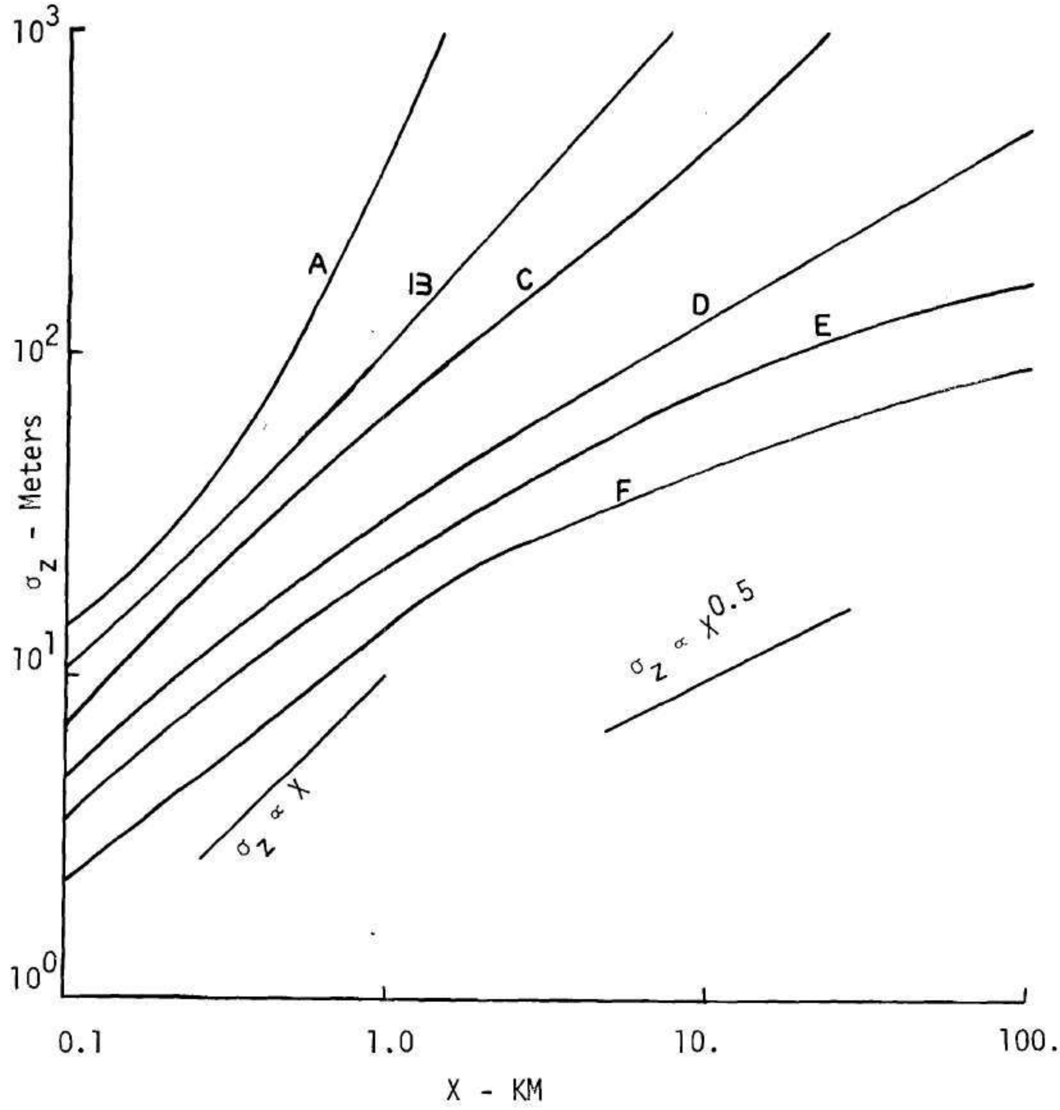


Figure 4. Vertical Dispersion Standard Deviations

plots were made for point sources rather than finite sources. Figure 4 represents the variation of σ_z with respect to downwind distance. It can be noticed that the bouyancy affects are significant in the determination of σ_z values. The trends of the plots at large downwind distance are different for different stability classifications. Here again, the initial portions of the plots have been experimentally found to be valid. There are not as many experimental verifications for the values at very large downwind distances.

The values of standard deviations for the fluctuating velocities σ_u, σ_v and σ_w are higher for an unstable atmosphere than those for a stable atmosphere. From equation (3.21) it can be seen that σ_z^2 is directly proportional to σ_w^2. This would mean that at all times the unstable stratification would give a greater extent of diffusion. The numerical values of the diffusion quantities σ_z and σ_y are accurate only within about a factor of two.

4.2 Estimation for the Value of ϵ

The value of energy dissipation rates depend on the energy cascading rate and also the rate at which energy is supplied at the large scale motions, both of which are approximately the same except for the dissipation by large scale motions. In order to study the variations on ϵ values at lower altitudes, the already existing functional relations for ϵ in the inertial subrange are used. From dimensional arguments it can be shown that

$$\epsilon \propto \sigma_{v}^{3}/\ell \qquad (4.1)$$

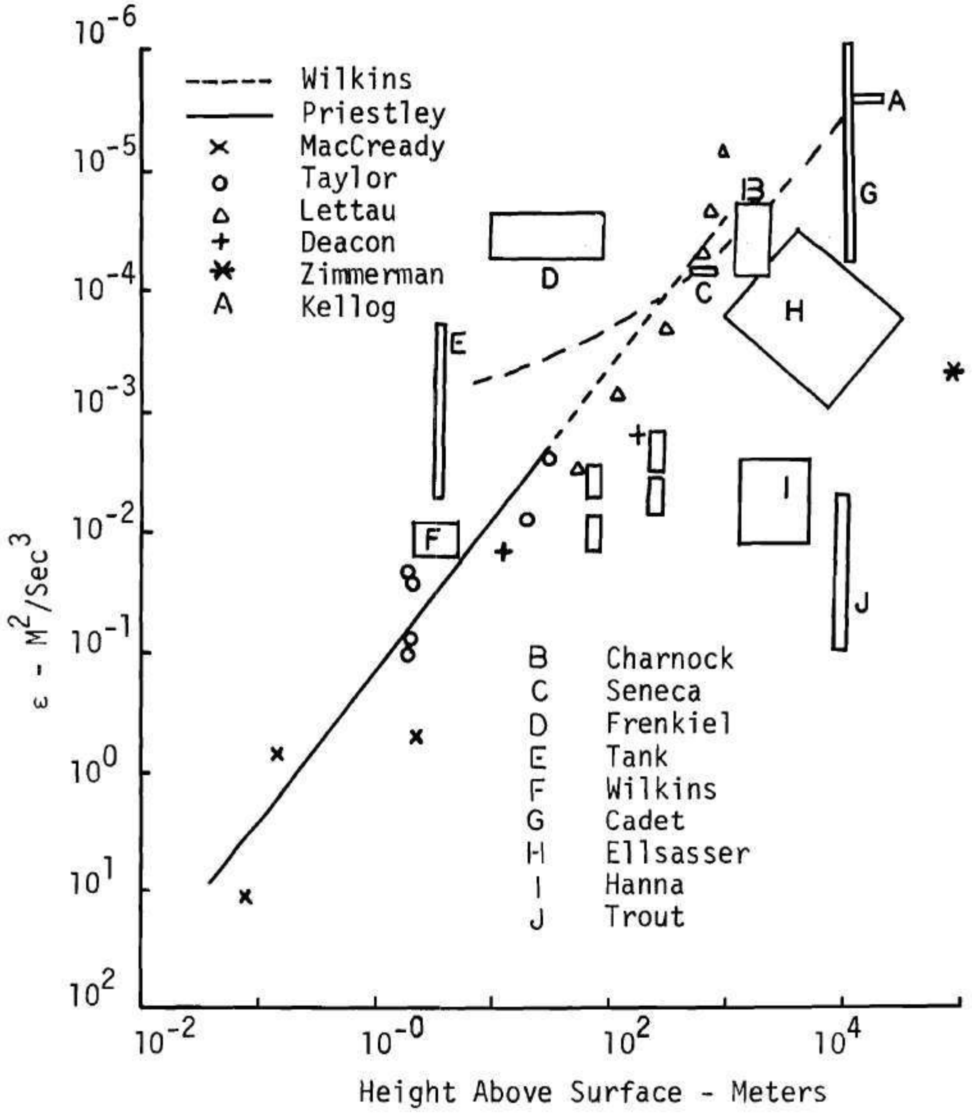


Figure 5. Rate of Energy Dissipation at Different Heights

where $\sigma_{\bar{v}}$ is the standard deviation for velocity fluctuations and ℓ is the Taylor integral scale. Equation (4.1) provides us with a means of estimating ϵ for a given $\sigma_{\bar{v}}$ at least within an order of magnitude. $\sigma_{\bar{v}}$ is a quantity which varies considerably with stratification. It is very high for unstable stratification and very low for stable stratification. For neutral stratification the value of ϵ for lower layers of the atmosphere is found to be about $10^{-3} m^2/sec^3$. The value of ϵ changes with altitude also. Figure 5 gives the vaule of ϵ as measured by different experimenters. There appears to be a wide scatter in data at higher altitudes. This is mainly because of the fact that turbulence at higher altitudes occurs in patches, and computations based on large range observations would give average values between patches of severe turbulence and patches of very small turbulence. Most of the recent measurements were made for evaluation of flight safety through severe turbulent patches. Hence, the values of ϵ so obtained will be much higher than those for the lower layers of the atmosphere. While considering atmospheric diffusion, the interest is on long range measurements. But measurements made in this area are very few and often data must be estimated from those for severe turbulence observations. The observations on ballon separations by Cadet (1976) are more suitable for higher altitudes than those obtained from others. This one data source itself shows a wide scatter in the results. Most often the choice of ϵ is made by observing how close a chosen value can predict theoretically the experimental results.

4.3 Estimation for the Value of k

This is a parameter which is introduced for the first time and hence there do not exist any experimental measurements so far. Out of the various types of heat transfer phenomena which exist in nature the appropriate type for modeling diffusion is that of natural convection. It has been observed in the case of heat transfer from solid surfaces that

$$h \propto (\Delta T)^{1/3} \tag{4.3}$$

where h is the heat transfer coefficient defined by the equation as shown below.

$$hA\Delta T = C\rho \ V \frac{dT}{dt} \tag{4.4}$$

Where C is the specific heat of air, ρ is the density, V is the volume of air under consideration, A the surface area of ambient air in contact with the moving air parcel. It can be seen that an estimate of k can be obtained in the following form.

$$k = \frac{hA}{\rho CV} \tag{4.5}$$

Before evaluating (4.5) it is necessary to specify the dimensions of the air parcels. The air parcels can be assumed to be masses of air which move together. In a turbulent motion particles excute random motions independent of each other. But in atmospheric motion the velocities are correlated and there will be a nonvanishing value of separation distance between particles for which the auto-correlation is almost unity. This particular dimension is best expressed by the Taylor microscale of turbulence. Substituting the empirical expressions given by Holman (1963) the following result can be obtained:

$$k = 0.0031 \frac{\left(-L_z \frac{dT}{dz}\right)}{\lambda} \tag{4.6}$$

where λ is the Taylor microscale. The value of a reference temperature difference between the air parcel and the ambient air is given by L_z dT/dz. Substitution of various values into (4.6) gives an estimate of k and for the atmosphere. It lies in the range of .01 to 0.1.

4.4 Stability A

Stability classification A corresponds to highly unstable stratification. Here the atmospheric lapse rate (-dT/dz) is such that a parcel of air which is moved up and down by the turbulent eddies finds itself at a temperature which accelerates the motion further. A parcel of air moving upwards will find itself at a temperature higher than the surrounding air and the bouyancy forces it upward and similarly a parcel of air which moves downward finds itself at a lower temperature than the surrounding air and bouyancy forces it still downward. The stability classification A has a characteristic velocity 2m/sec and a lapse rate of 0.019°/m. The initial portions of Figures 3 and 4 are drawn such that a t^2 variation is followed. The representative values for σ_u, σ_v and σ_w can be hence obtained from one point on the plot using the equations below.

$$\sigma_z = \sigma_w t = \sigma_w X_o | \bar{U}$$

and

$$\sigma_y = \sigma_v t = \sigma_v X_o | \bar{U}$$

where X_o is the downwind distance at the first point on the plot and $\bar{U}$

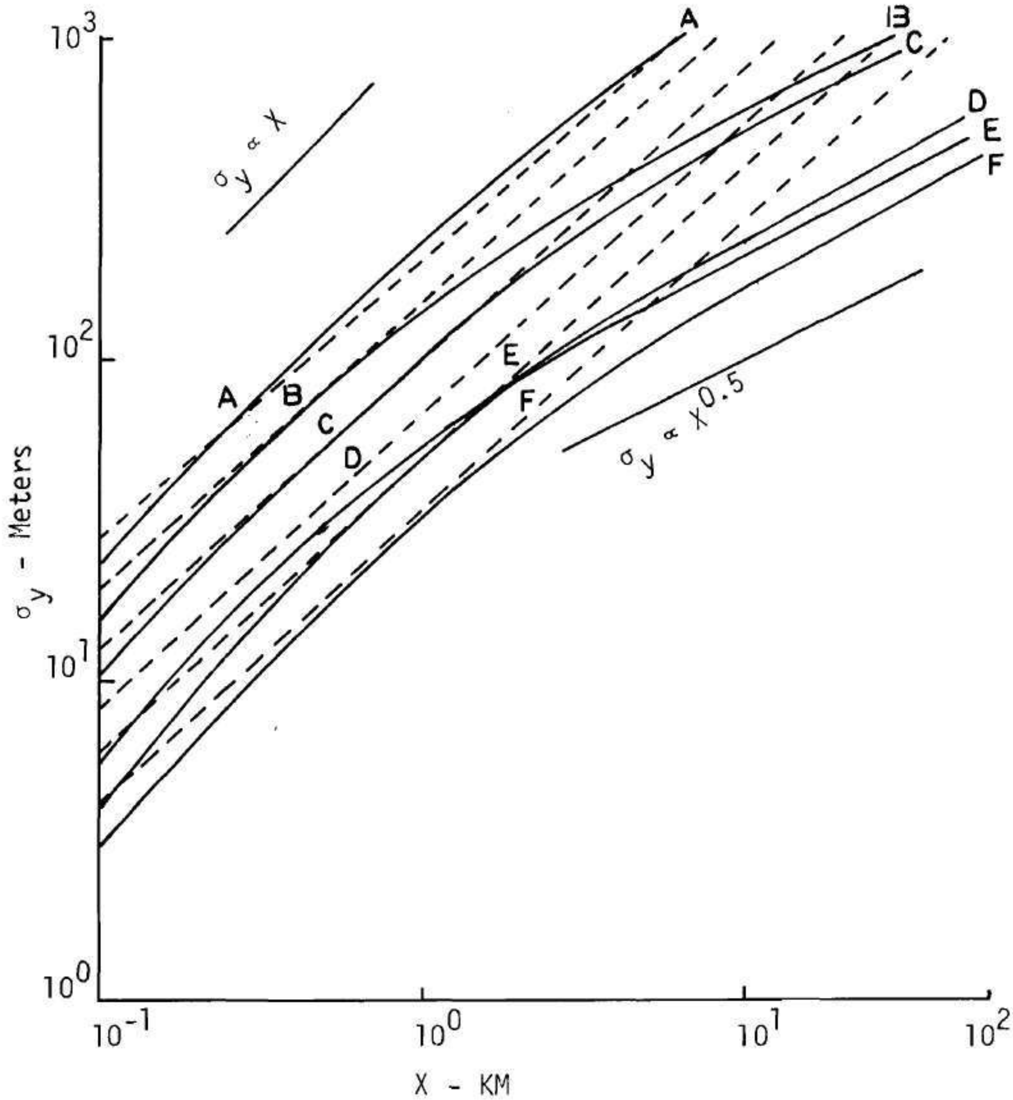


Figure 6. Horizontal Dispertion Standard Deviation, Present Model Against Experiments.

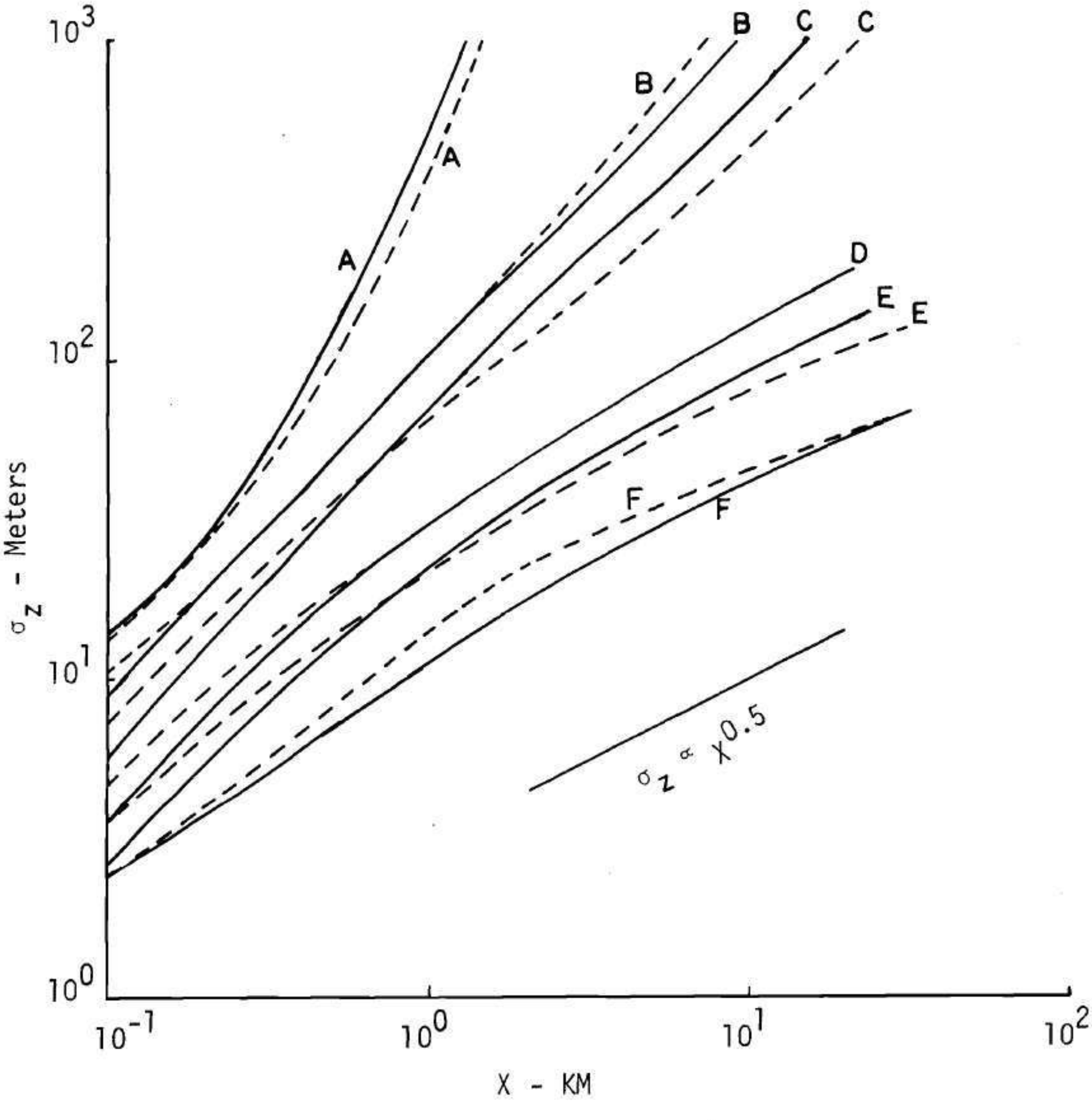


Figure 7. Vertical Dispertion Standard Deviation, Present Model Against Experiments.

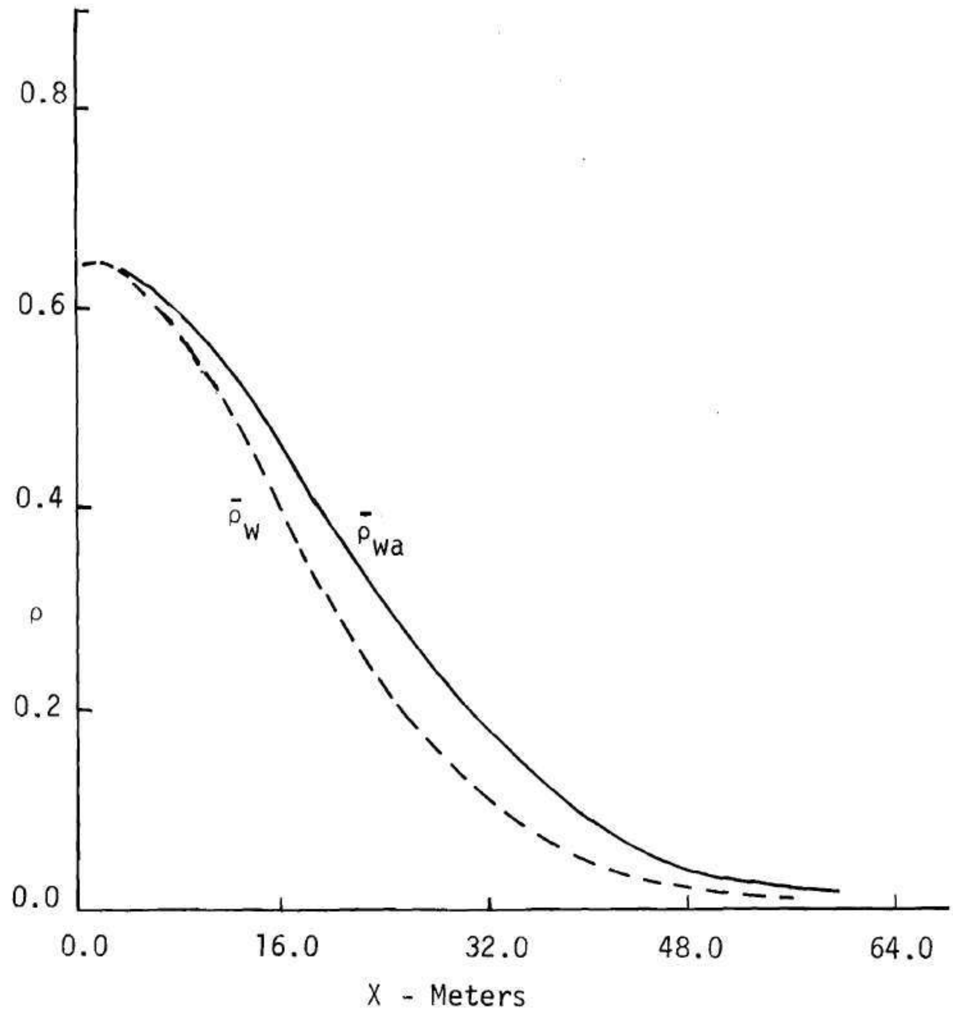


Figure 8. Variation of $\bar{\rho}_w$, Stability A

is the mean velocity. For stability A the above procedure gives the following results.

$$\sigma_u = \sigma_v = 0.55\text{m/sec}$$

$$\sigma_w = 0.286\text{m/sec}$$

The initial value of the cloud was taken to be 2m in size. This was, again, chosen so that the initial point and the present model solution are as close as possible. The choices of ϵ and k are made so that the best fit is obtained within the accuracy of ϵ and k. It was found that

$$\epsilon = 1 \times 10^{-3}\text{m}^2/\text{sec}^3$$

$$k = 0.04/\text{sec}$$

It can be observed that the variation of σ_y becomes linear with respect to time at great downwind distances. The variation of σ_z is exponential as was observed from the analytical solution itself. The temporal changes are predicted remarkably well with these parameters. Figures 6 and 7 show these variations. Fig. 8 shows the variation of $\bar{\rho}_w$ with time and also the variation of an approximation by the functional form given by equation (3.34). It shows that a parameter α does exist independent of time for modeling $\bar{\rho}_w$. Figure 14 shows the effect of wind shear on the diffusion. The wind shear was calculated using the formula obtained by Justus and Mikhail (1976) as shown below.

$$\frac{V_1}{V_2} = \left(\frac{Z_1}{Z_2}\right)^n \tag{4.7}$$

Where V_1 and V_2 are wind velocities at heights Z_1 and Z_2 the value of n

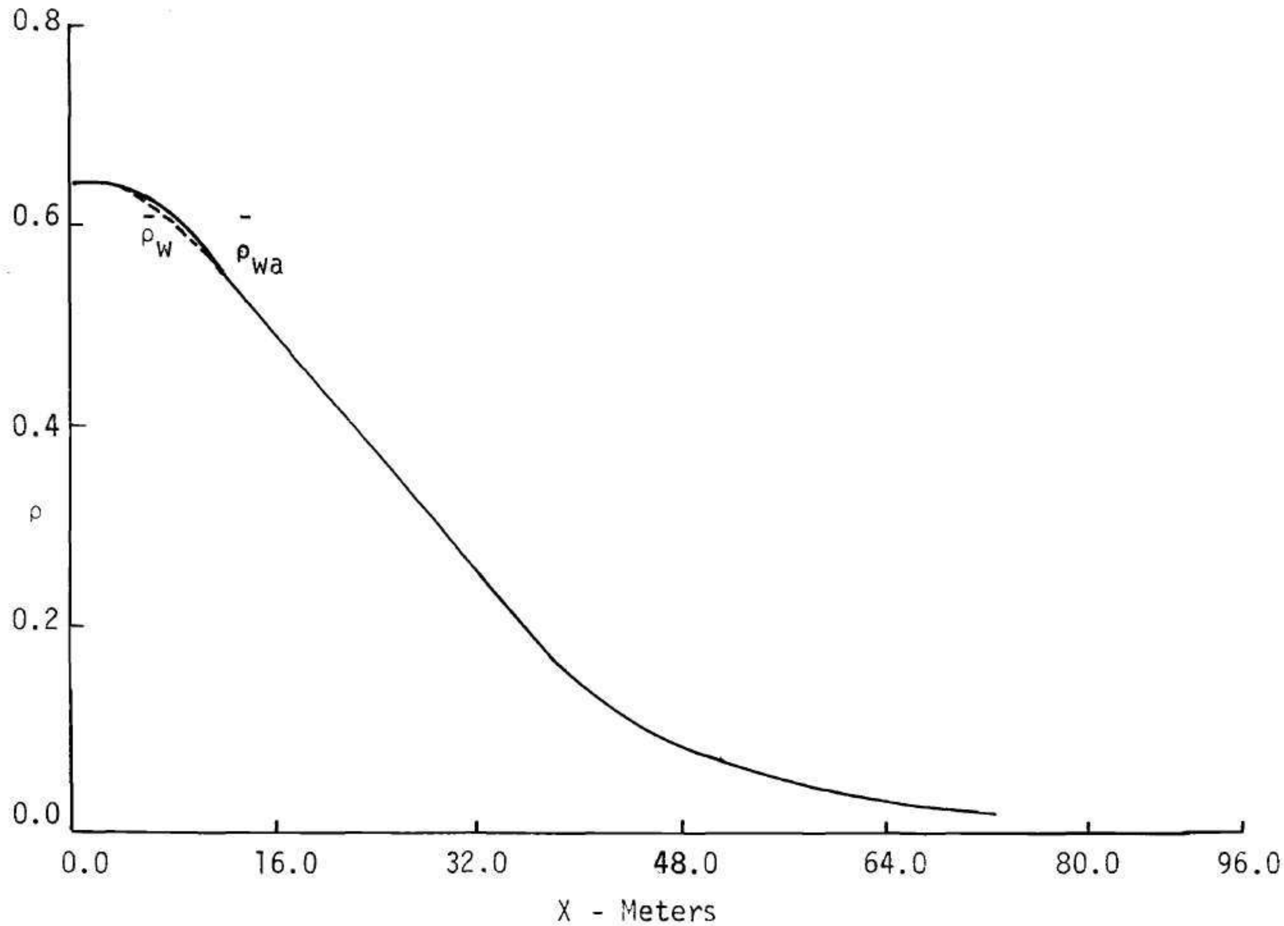


Figure 9. Variation of $\bar{\rho}_W$, Stability B

for the stability A was 0.21.

4.5 Stability B

Stability classification B corresponds to unstable situation again. But the characteristic of B stability is markedly different than stability A. The values of σ_u, σ_v, and σ_w are estimated in the same way as was done before. The values obtained are

$$\sigma_u = \sigma_v = 0.76\text{m/sec}$$

$$\sigma_w = 0.432\text{m/sec}$$

The mean wind for B stability is estimated as 4m/sec. The length scales are 10m,10m and 5m in the x, y and z directions respectively. The initial size of the diffusing cloud was assumed to be 2m as before. The value of ϵ and k are the following

$$\epsilon = 1 \times 10^{-2} m^2/\text{sec}^3$$

$$k = 0.066/\text{sec}$$

the lapse rate is 0.017. It can be seen that the exponential nature of the σ_z variation is damped to some extent and σ_z does not increase as fast as it did for the case of stability A. The variation of σ_y^2 again shows a t variation at large downwind distances as was predicted by the analytical solution. Figures 6 and 7 shows the variation of σ_y and σ_z with downwind distances. Figure 9 again shows that the variation of $\bar{\rho}_w$ can be approximated as given by equation (3.34). Figure 14 shows the effect of wind shear on diffusion. The value of n for stability B was 0.19.

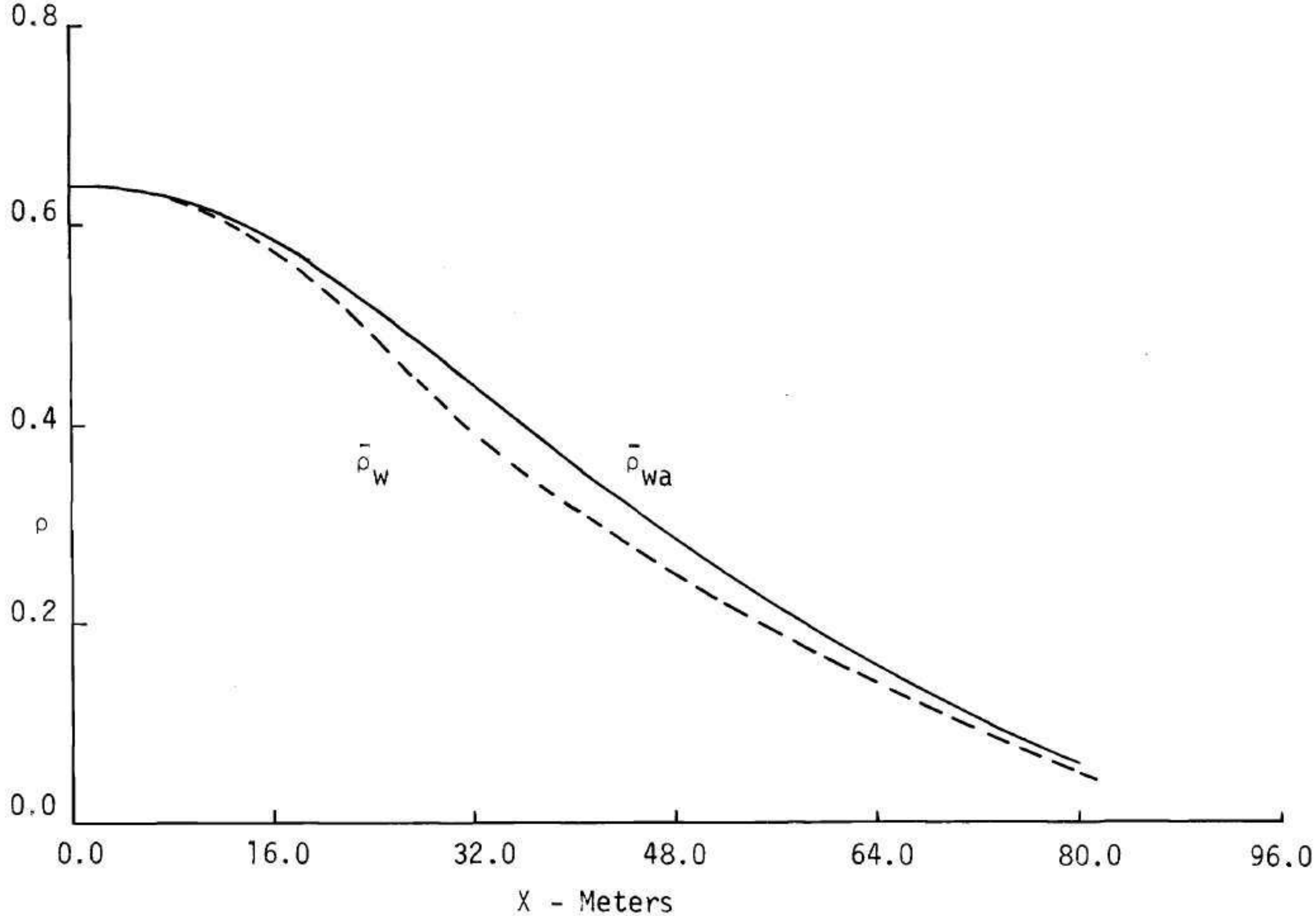


Figure 10. Variation of $\bar{\rho}_w$, Stability C

4.6 Stability C

Stability classification C again is unstable stratification. But the degree of instability is much less as compared to A and B. The values of σ_u, σ_v and σ_w are again estimated using the first point on the experimental plot.

$$\sigma_u = \sigma_v = 0.63\text{m/sec}$$

$$\sigma_w = 0.375\text{m/sec}$$

The representative mean wind for stability C is 5m/sec and the length scales in the x, y and z directions are 10m, 10m and 5m respectively. The initial size of the cloud is 2m. The values of ε and k are the following

$$\varepsilon = 5 \times 10^{-3} \text{m}^2/\text{sec}^3$$

$$k = 0.076/\text{sec}$$

It can be seen that the exponential nature is completely removed and the vertical diffusion almost follows a t^2 variation. The variation of σ_y^2 is like t^2 during the initial periods and at large downwind distances this becomes proportional to t. Figures 6 and 7 show the variation of σ_y and σ_z with downwind distance. Figure 10 shows the variation of $\bar{\rho}_w$ as against that obtained by equation (3.13). Figure 14 shows the effect of wind shear on diffusion. The results of the earlier model were far removed from those obtained by the present model. The value of n for stability C was chosen to be 0.156.

4.7 Stability D

The stability classification D is the neutral stratification. A parcel of air when moved up and down by the turbulent fluctuation undergoes adiabatic changes when stratification is neutral. The ambient air which surrounds the parcel of air has a lapse rate which is equal to the dry adiabatic rate. Hence the moving parcel of air always has the temperature of the ambient air it comes into contact with. The bouyancy forces do not play any part in the diffusion. The diffusion process in the vertical direction will be the same as that in the horizontal except for the wind shear effects. The values of rms fluctuation velocities are the following.

$$\sigma_u = \sigma_v = 0.4\text{m/sec}$$

$$\sigma_w = 0.235\text{m/sec}$$

The characteristic wind velocity for this stability is 5m/sec. The length scales are chosen to be 10m, 10m and 5m respectively for L_x, L_y and L_z. Initial size of the cloud is taken to be 2m. The value of ϵ is found to be

$$\epsilon = 5 \times 10^{-3} m^2/\sec^3$$

the value of k for this particular situation is not important because there is no heat transfer between the air parcel and the atmosphere. Hence the value of k does not play any part in the process of diffusion. The variation of σ_y and σ_z are shown in Figures 6 and 7. It can be

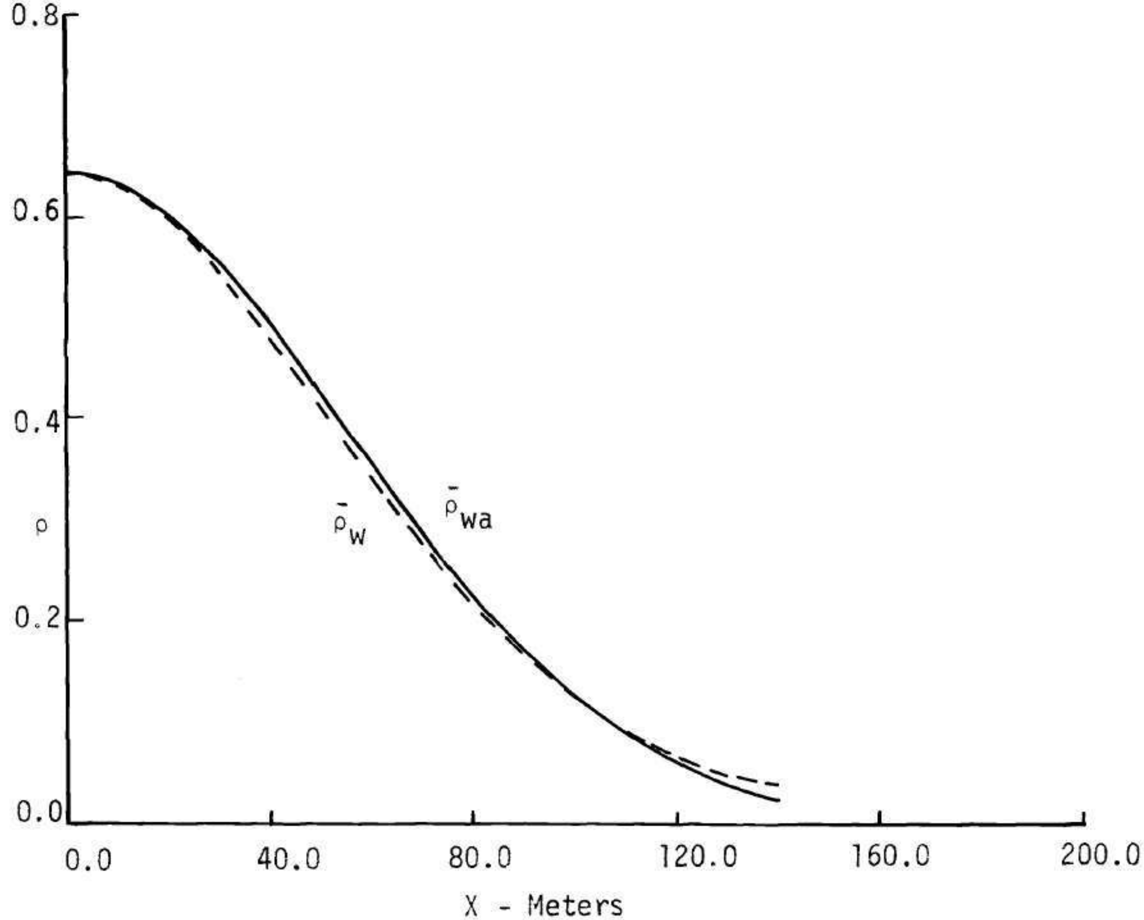


Figure 11. Variation of $\bar{\rho}_w$, Stability D

seen that at large values of downwind distances σ_z^2 follows a t variation. The variation of σ_y with x follows the Pasquill plot until it becomes proportional to $x^{1/2}$. Figure 11 shows the variation of $\bar{\rho}_w$ with time and the variation given by (3.13). Figure 14 gives the effect of wind shear. The value of n was taken to be 0.175.

4.8 Stability E

The stability E characterizes a stable atmosphere. Here the value of Γ is positive and the bouyancy effects are opposite to that of unstable atmosphere. The parcel of air which executes random up and down motion comes in contact with outside air which has temperatures such that the motions are damped out. For example a gas parcel which moves dry adiabatically upwards will encounter an outside air mass which is at a higher temperature but at the same pressure. The bouyancy effects will be to push the parcel down thereby damping the upward motion. On the other hand a parcel of air which moves downward will find that the surrounding air is colder than itself and bouyancy forces will tend to move it upward. As was seen from the analytical solution the large time behavior depends on the value of k if ωk is of the same order of magnitude as $2g\Gamma$. The r.m.s. velocity fluctuations for this stability class are

$$\sigma_u = \sigma_v = 0.186\text{m/sec}$$

$$\sigma_w = 0.107\text{m/sec}$$

the characteristic wind velocity for this stability is 3m/sec. The length scales in the X, Y and Z directions are 10m, 10m and 5m respectively. The initial size of the cloud was chosen to be 2m. The values

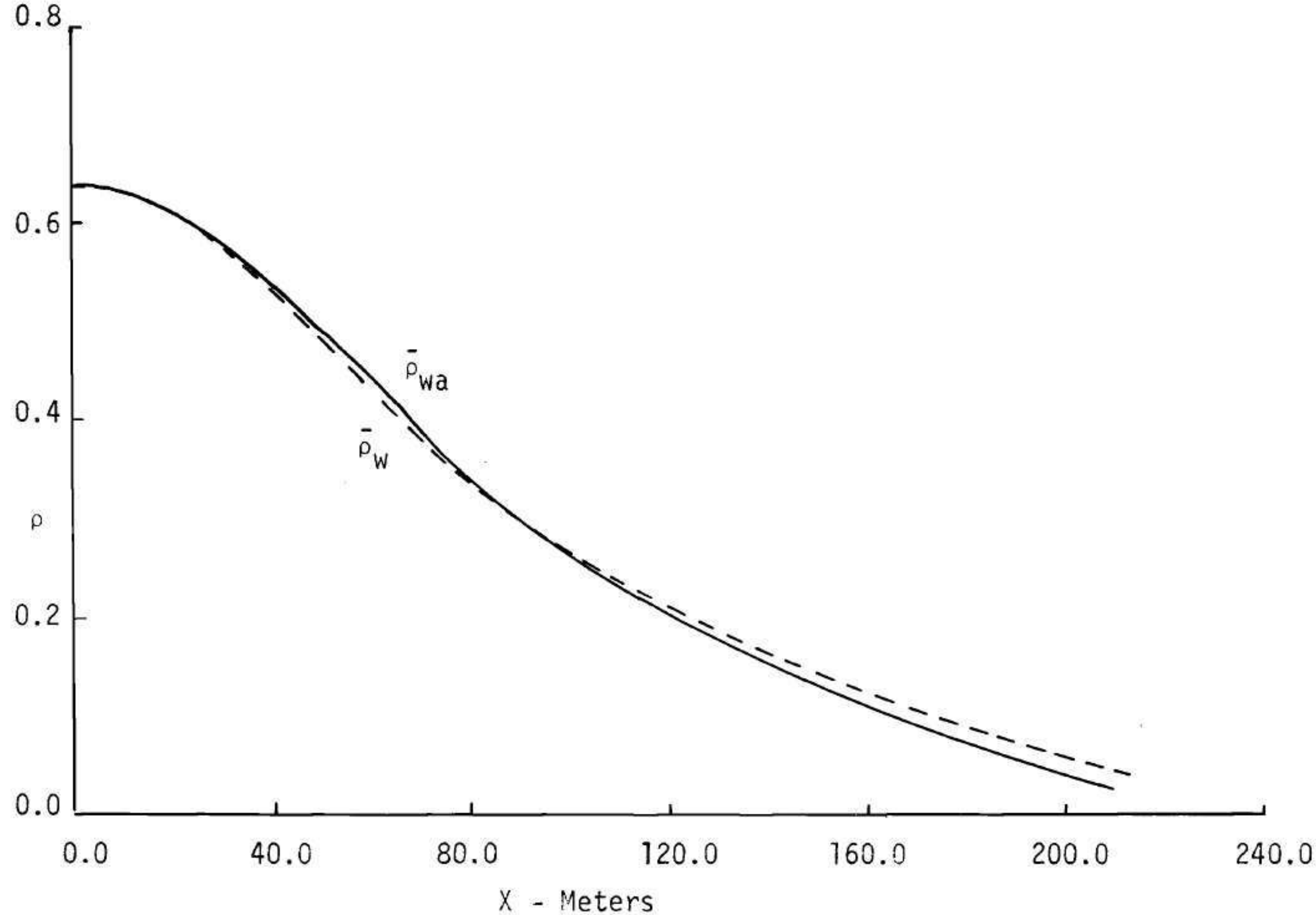


Figure 12. Variation of $\bar{\rho}_w$, Stability E

of ϵ and k are the following

$$\epsilon = 3 \times 10^{-4} m^2/sec^3$$

$$k = 0.092/sec$$

The large time behavior follows a linear relation with the downwind distance, as was expected from the analytical solution. Figures 6 and 7 show the variation of σ_y and σ_z with downwind distances. It is found that the earlier model was not suited for stable stratifications. The present model is able to predict the trend very accurately. Figure 12 is again drawn to show the variation of $\bar{\rho}_w$ and its dependence on a parameter α. Figure 15 shows the variation of 1/2 σ_{zx} and 1/2 σ_{zy} with the downwind distance. The value of n was 0.215.

4.9 Stability F

Stability classification F characterizes an extreme stable stratification. Here the value of Γ is positive. The turbulence is very low, because the stability damps out all vertical motions. The r.m.s. wind velocities are found to be

$$\sigma_u = \sigma_v = 0.08 m/sec$$

$$\sigma_w = 0.126/sec$$

Figures 6 and 7 show the variation of σ_y and σ_z with downwind distance. The earlier model by Justus and Mani was not effective for this stability. The new model is able to predict the trend very well. At large values of X σ_z^2 again follows a linear dependence on X. Figure 13 is drawn to show the variation of ρ_w on a parameter α. Figure 15 shows the effect of wind shear on diffusion. The value of n was 0.412.

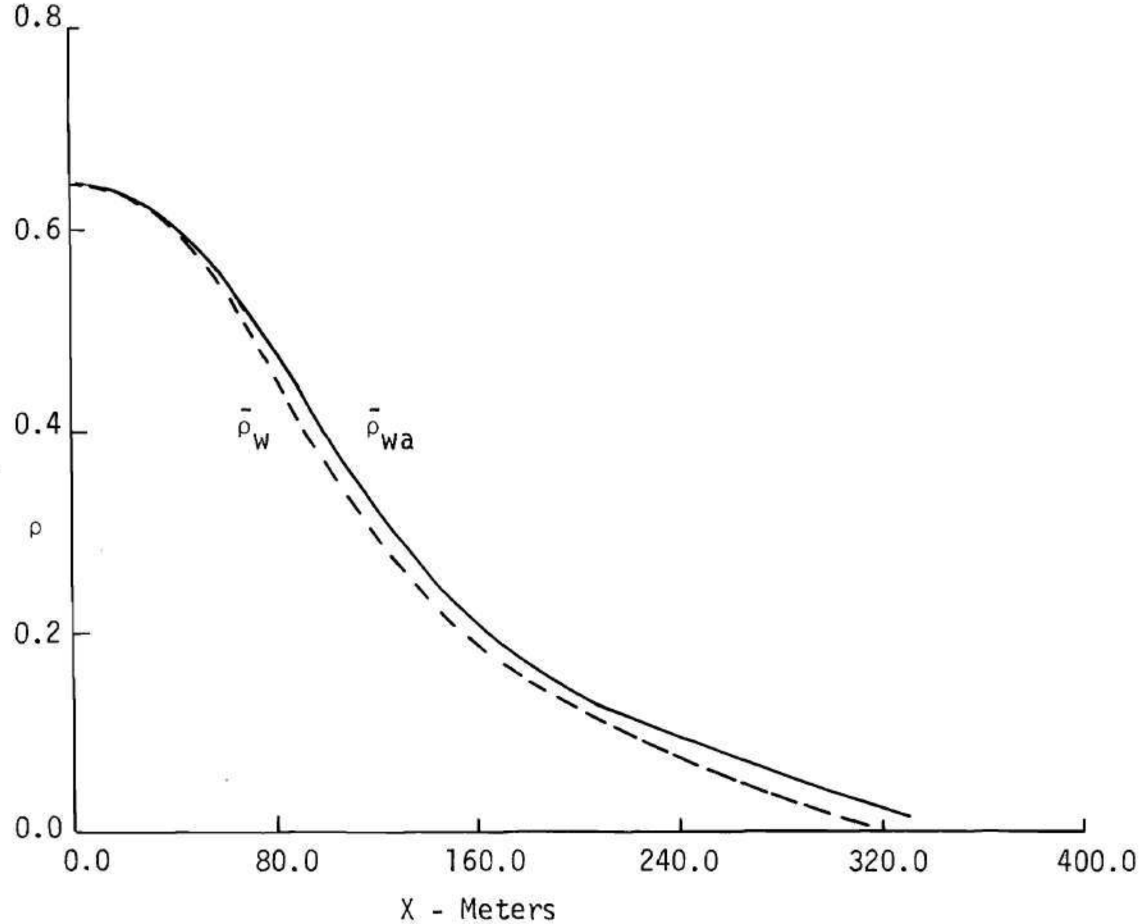


Figure 13. Variation of $\bar{\rho}_w$, Stability F.

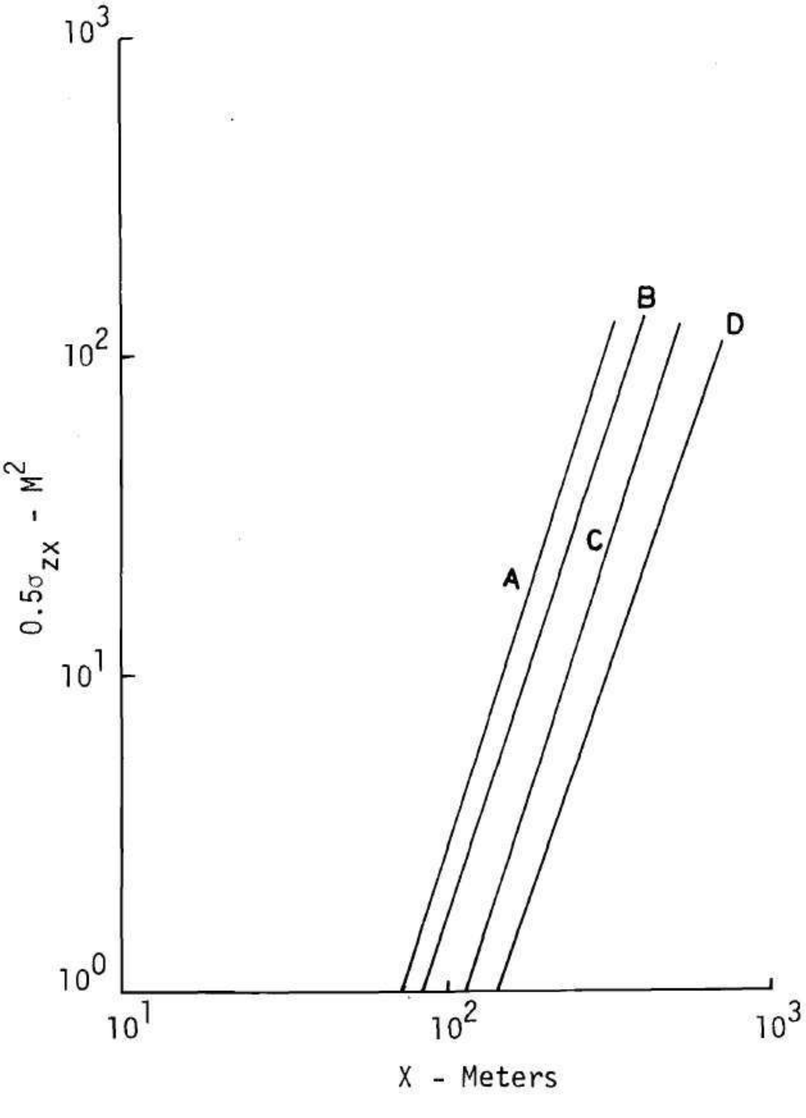


Figure 14. Variation of σ_{zx} with Downwind Distances for Stabilities A,B,C and D.

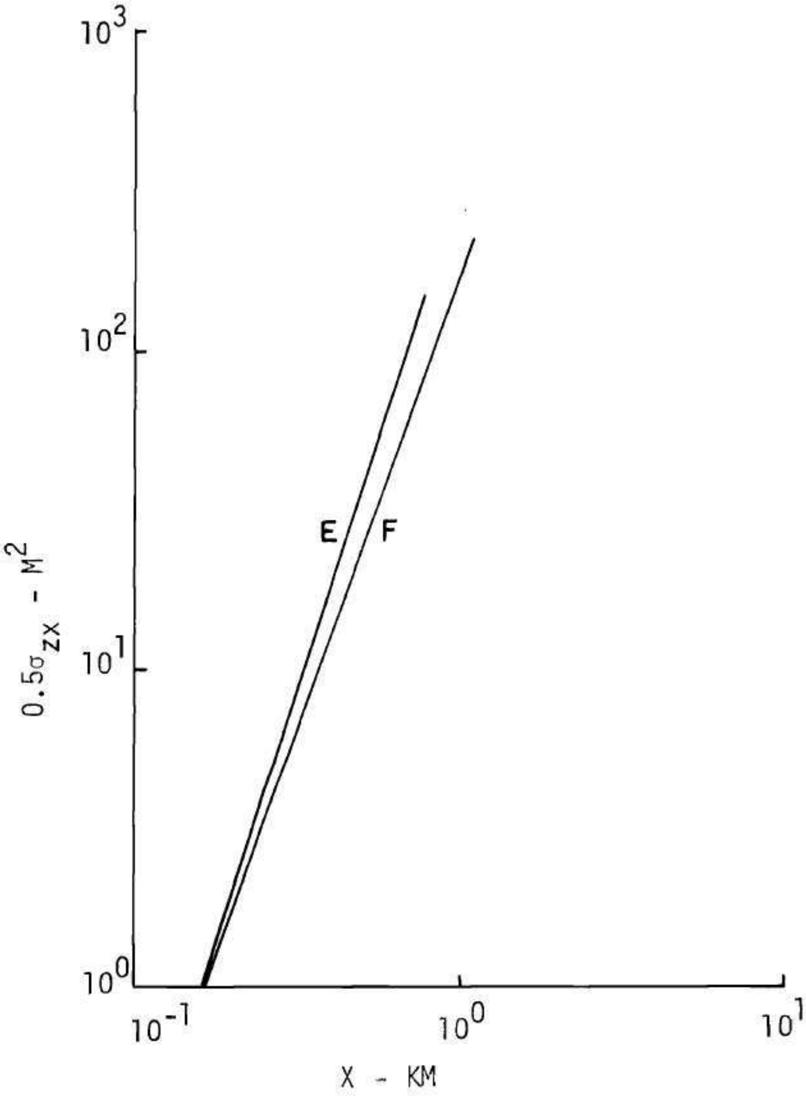


Figure 15. Variation of σ_{zx} with Downwind Distances for Stabilities E and F.

TABLE 2: Values of ϵ and k for Various Stabilities

Stability	ϵ m^2/sec^3	k /sec
A	1×10^{-3}	0.04
B	1×10^{-2}	0.066
C	5×10^{-3}	0.076
D	5×10^{-3}	—
E	3×10^{-4}	0.092
F	3×10^{-5}	0.126

TABLE 3: Values of α, σ_u, σ_v, σ_w and Γ for Various Stabilities

STABILITY	A	B	C	D	E	F
α/sec^2	.005	.015	.009	.00403	.0009	.00019
σ_u m/sec	0.55	0.76	0.63	0.4	0.186	0.08
σ_v m/sec	0.55	0.76	0.63	0.4	0.186	0.08
σ_w m/sec	0.286	0.432	0.375	0.235	0.107	0.047
$\Gamma \times 10^5$ sec^{-1}	-3.3	-2.5	-1.8	0.0	2.1	7.1

Table 2 summarizes the values of ϵ and k used for all the stability conditions. Table 3 shows the dependence of the values of α on the various parameters determining the stability conditions. Figures 14 and 15 are plotted separately because of the fact that the values of σ_{zx} for E and D stabilities are too close together.

CHAPTER V

STRATOSPHERIC DIFFUSION

5.1 Diffusion at Higher Altitudes

As was mentioned in Chapter I the motions in the stratosphere consist of very large scales. Most of these wave systems which exist in the stratosphere travel around the earth. The stratosphere is characterized mostly by a stable stratification. Hence the vertical motions are greatly inhibited by the bouyancy forces. The horizontal motions will contain patches of turbulence and regions of laminar flow. Any pollutant that is introduced into the stratosphere travels around the earth much faster than in the case of troposphere. In the troposphere the three distinct cells of motion inhibits pollutants from moving from one cell to the other. Whereas in the stratosphere only one cell exists and the pollutants can easily travel meridionally to a large extent.

The measurements of stratospheric diffusion are made in zonal bands and the measurements in one band are averaged together for documentation. The large scale motion does not cause any relative motion between the particles of the diffusing cloud. But when zonal averaging is done these large scale motions introduce an apparent diffusion which is characteristic of large scale turbulence. While comparing the stratospheric diffusion measurements as against the theoretical model the scales of motion will be the large scale motion rather than the Taylor integral scales. The stratospheric diffusion measurements compiled by

Bauer (1974) are used in the present comparison.

In order to make a theoretical prediction at stratospheric heights, information regarding appropriate turbulence statistics is required. This is obtained from the Global Reference Atmospheric Model.

5.2 Global Reference Atmospheric Model (GRAM)

GRAM is a computer model developed by Justus, Woodrum, Roper and Smith (1974) for predicting mean motions, and atmospheric turbulence in a statistical sense. This program can predict atmospheric properties and wind statistics at a given latitude and longitude for a height range of 0 to 110km, and also for different times of the year. This program is capable of predicting quasi-biennial oscillations and random perturbations to simulate partially the variability due to synoptic, diurnal, planetary wave and gravity wave variations. This program is an amalgamation of three different models viz. the NASA four dimensional world wide model, the Groves height and latitude dependent monthly means model and the Jachia 1970 model. The above three sections are incorporated in order to compute the various parameters up to 110km altitude.

5.3 GRAM-DIFFUSION model

The GRAM-DIFFUSION model is a combination of the GRAM program and the diffusion program. Thus given an initial cloud size and its location in longitude and latitude and height above the surface along with the time of the year the program computes the trajectory of the diffusing cloud and also the diffusion related parameters. This model is more suitable for stratospheric studies rather than tropospheric studies mainly because of the large magnitudes of the convecting motions of the strato-

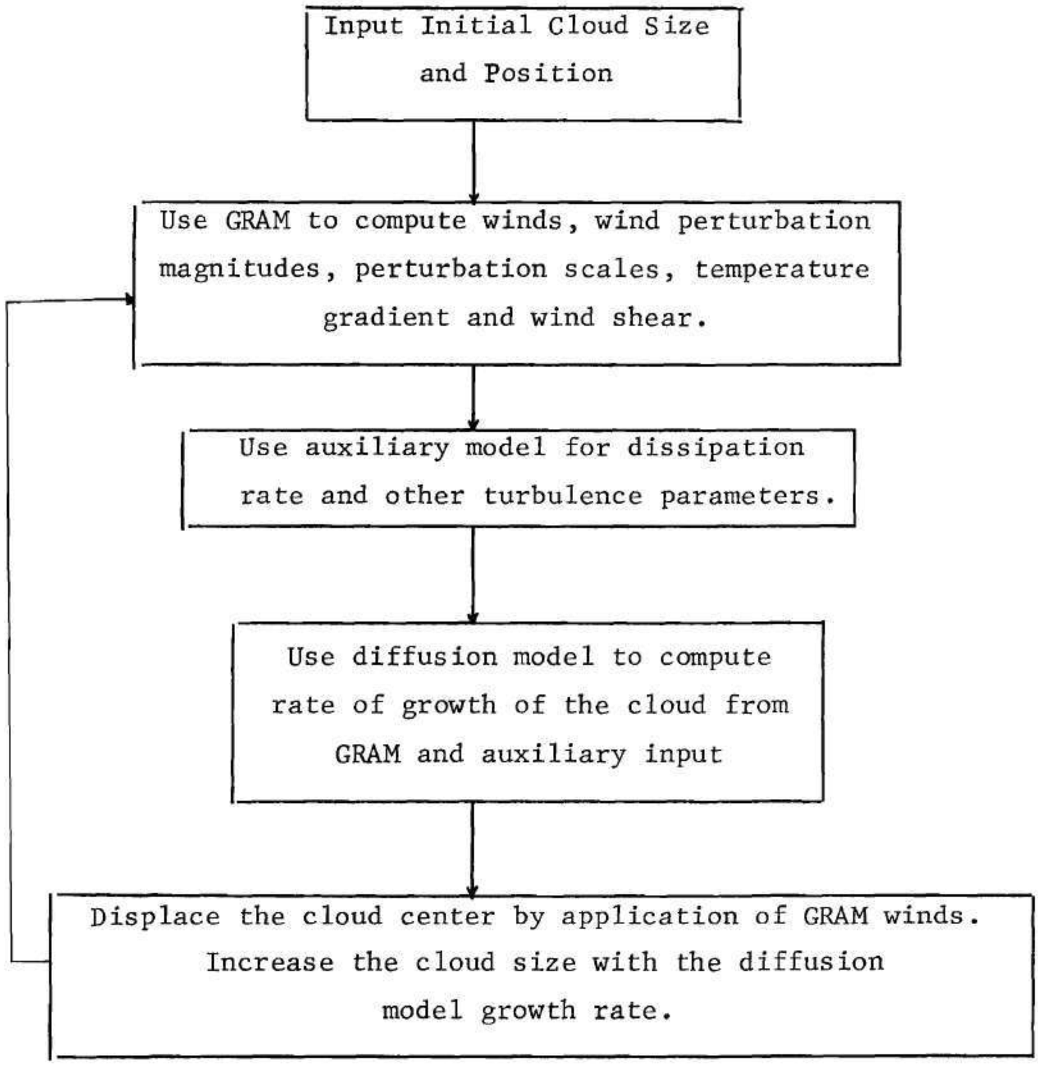


FIGURE 16: Flow Chart for the GRAM-DIFFUSION Model.

TABLE 4: The Vertical Turbulent Velocity Fluctuation

Lower Atmosphere		Stratosphere		Upper Atmosphere	
Altitude Meters	σ_w m/sec	Altitude Meters	σ_w	Altitude km	σ_w m/sec
200-600	0.45				
600-1200	0.30			90-100	14.00
1200-1600	0.31			100-110	15.89
1600-2000	0.45	Stratospheric means	0.30		
2000-2500	0.40				
2500-3000	0.33				
3000-4000	0.31				
4000-5000	0.35				
5000-6000	0.30				
6000-7000	0.37				
7000-8000	0.36				
8000-9000	0.27				
Balloon Data		Lilly, Waco & Adelfang (1974)		Justus (1969)	

sphere. The GRAM program has the capability of dividing the atmospheric motion into small scale and large scale components. The small scale motions are mainly the turbulent motions and the large scale motions are more steady in nature.

The flow chart showing the functioning of the GRAM-DIFFUSION model is presented in Figure 16. Given the initial size and the location of the diffusing cloud the GRAM program extracts the measured values of the thermodynamic properties and standard deviations from data tapes for grid locations around the given location. The values for the given location are then interpolated from the grid point values. The mean winds are calculated using geostrophic balance conditions. The GRAM program does not have the capability to predict the value of σ_w. Values for σ_w for various heights appear in Table 4. Instead of reading this table of values every time an empirical relation in the following form is used.

$$\sigma_w^2 = \lambda_1\left(\sigma_u^2 + \sigma_v^2\right) \tag{5.1}$$

And most often a value of 0.1 for λ_1 gives good results.

5.4 Diffusion Computation

Diffusion calculations were conducted for heights of 32km and 40km during the month of January and July. The results are plotted against the results of Bauer in Figures 17 and 18. The value of ϵ chosen was $10^{-5}m^2/sec^3$ and k was 0.126. Figure 17 shows the variation of horizontal spread with time. This compares very well with the results of Hage et al (1966). Figure 18 shows the vertical diffusion and it can be seen that the vertical spreading is very much smaller than the

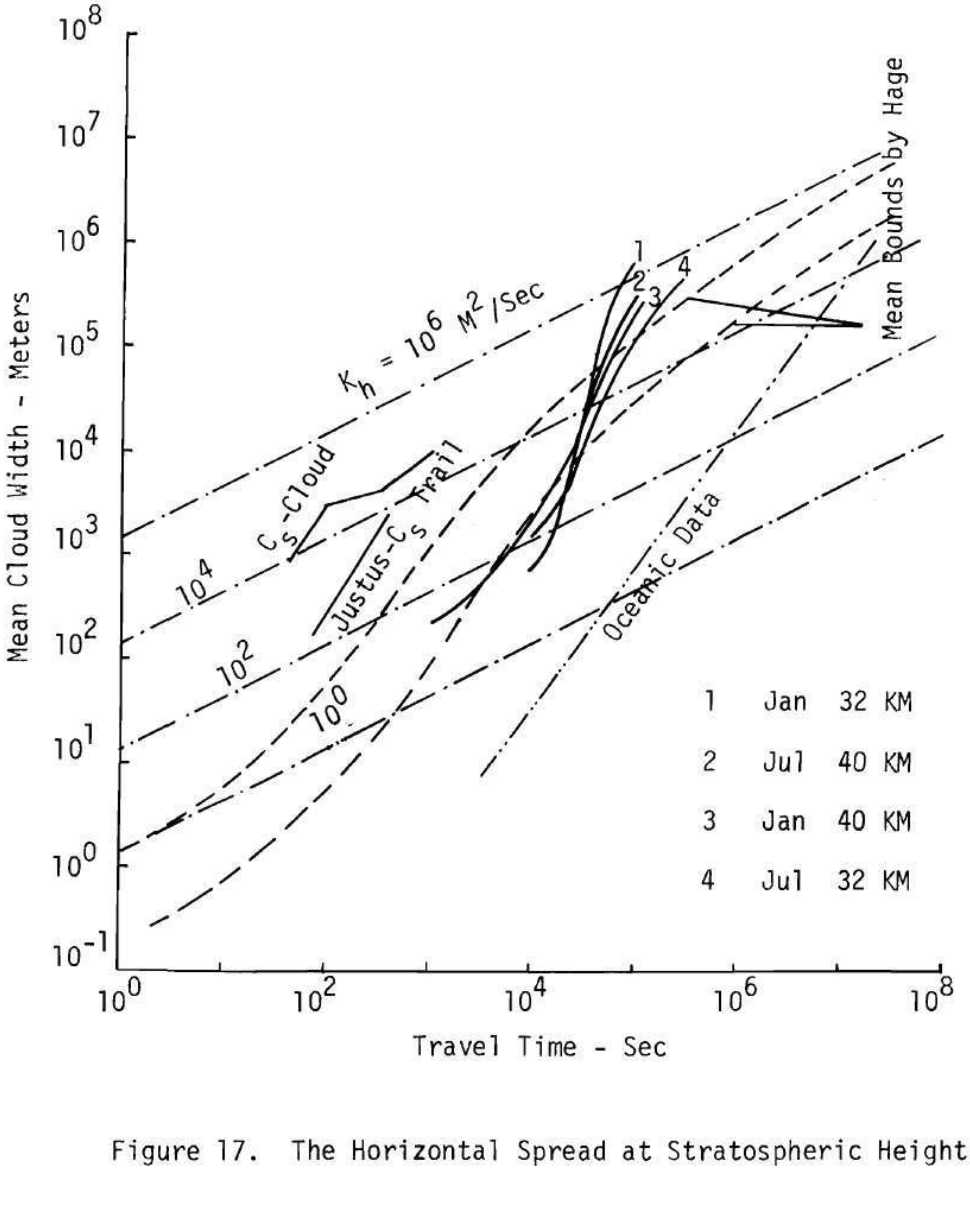


Figure 17. The Horizontal Spread at Stratospheric Heights.

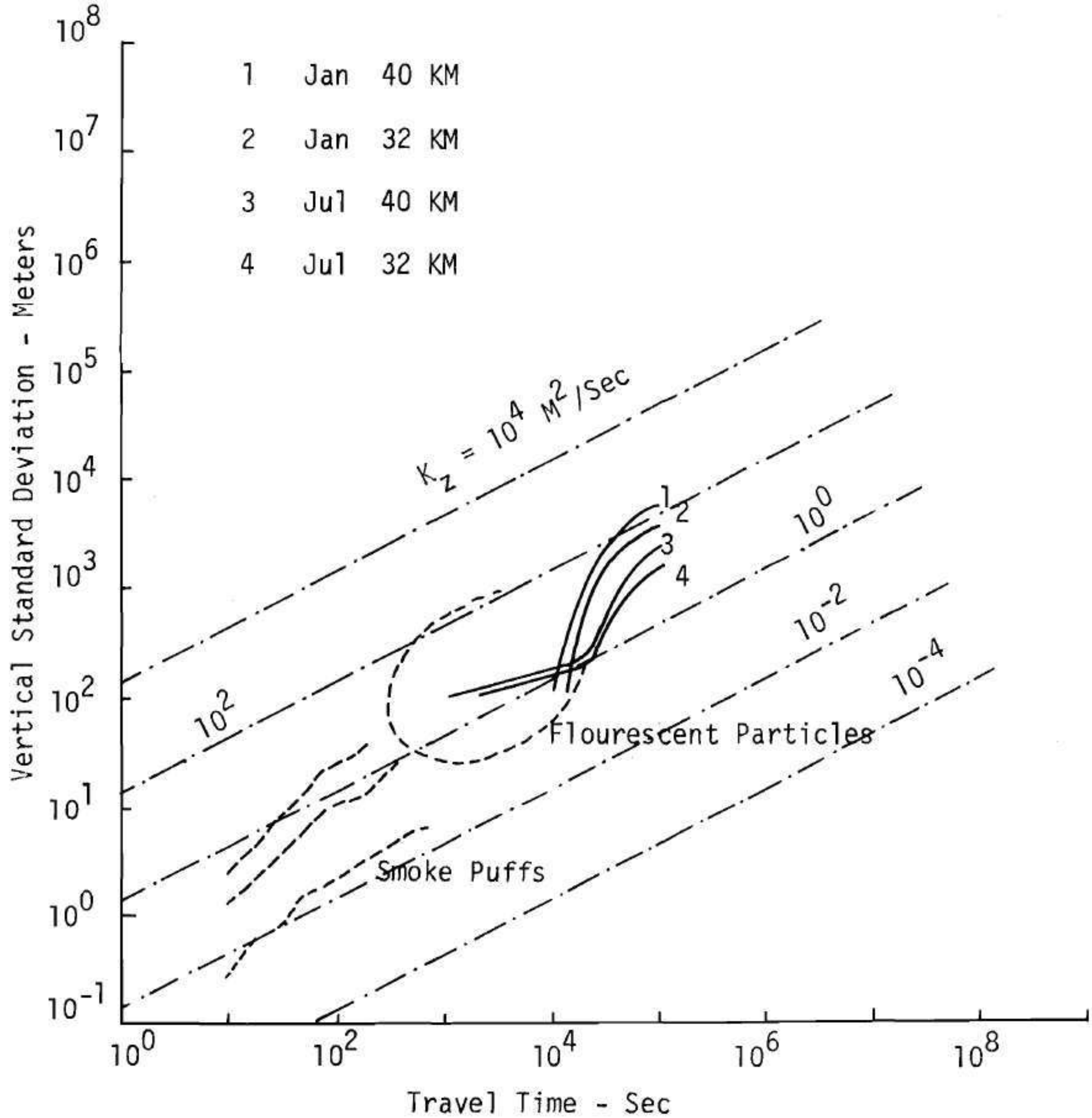


Figure 18. Vertical Spread at Stratospheric Heights.

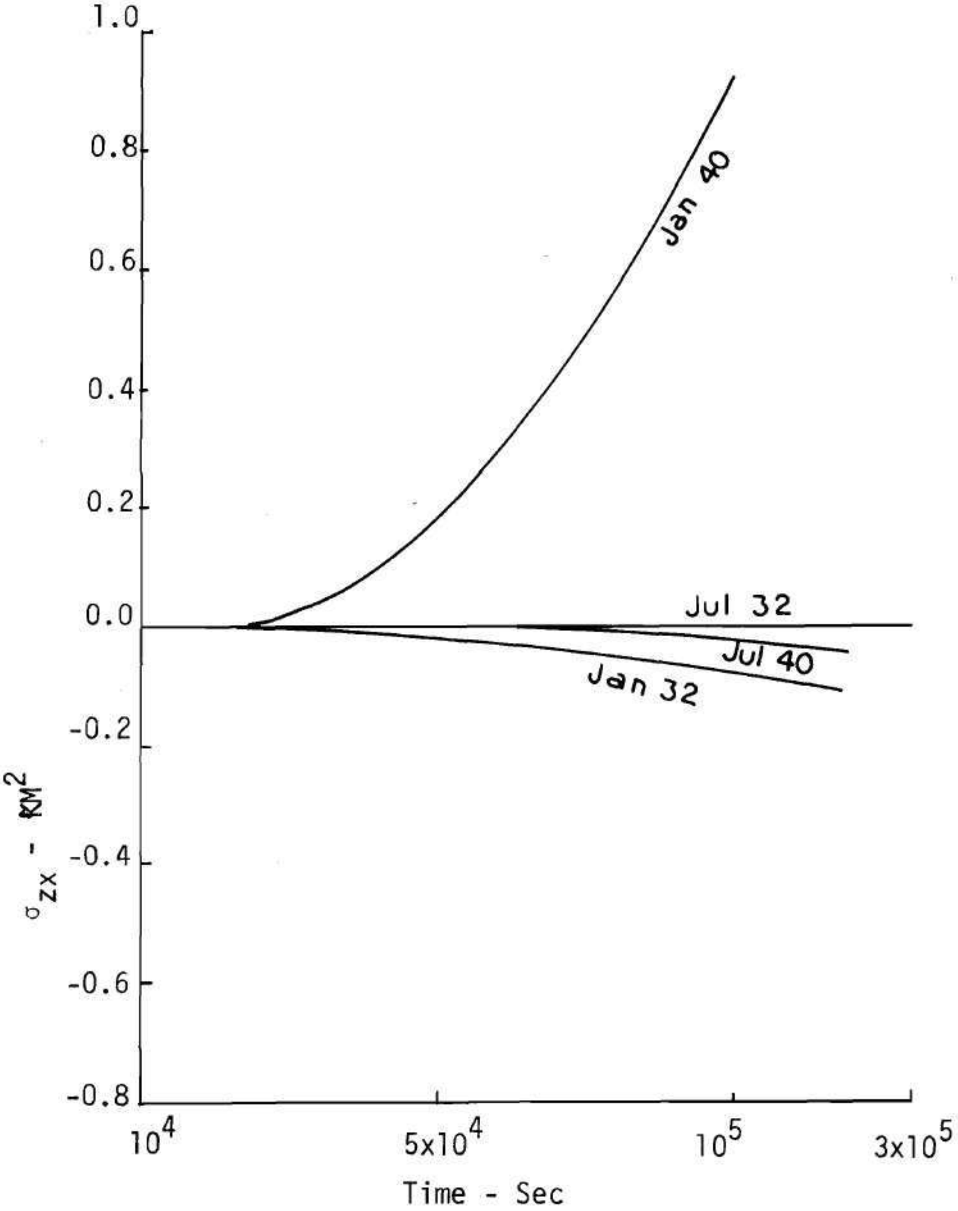


Figure 19. Variation of σ_{zx} at Stratospheric Heights.

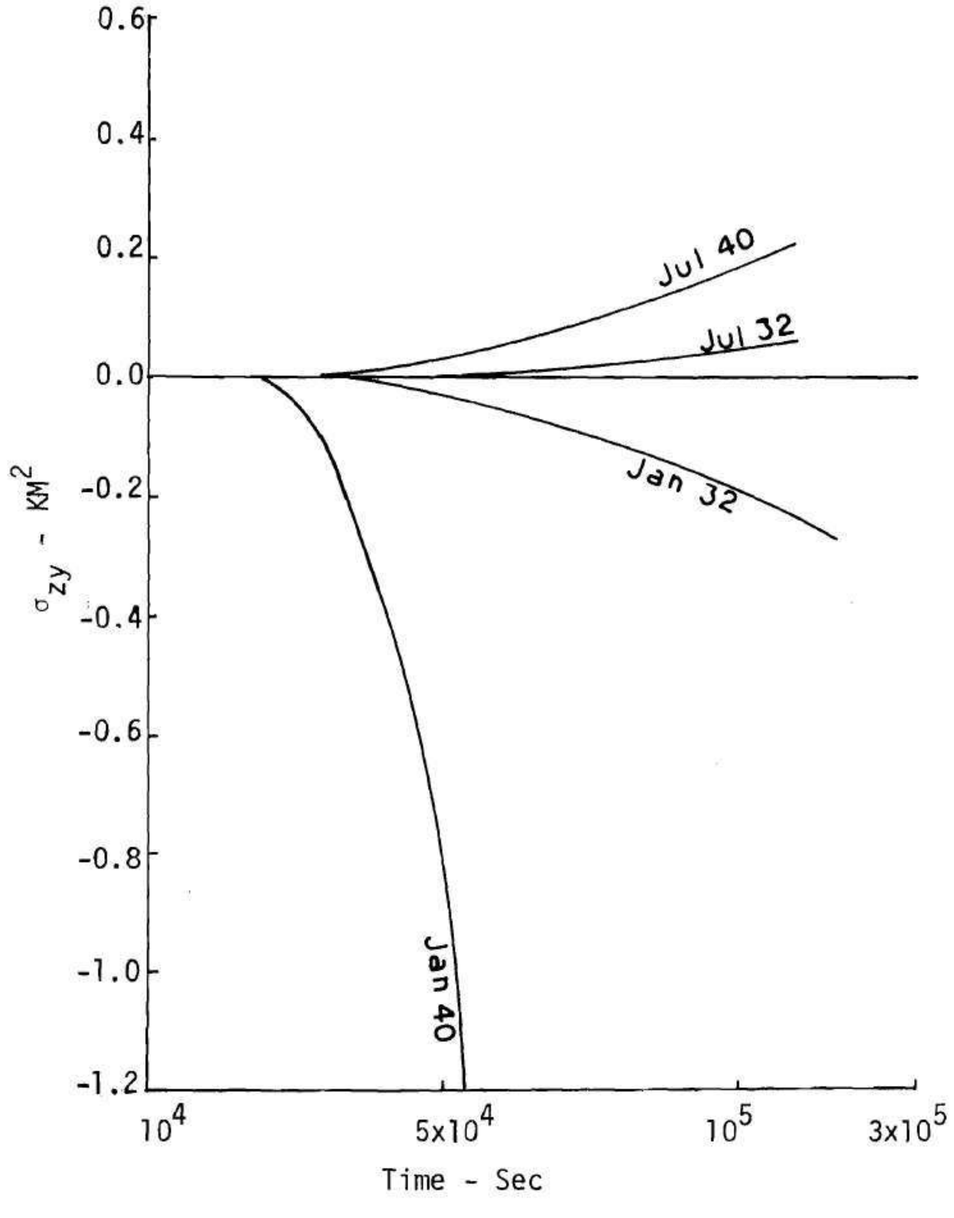


Figure 20. Variation of σ_{zy} at Stratospheric Heights.

horizontal spreading. Figure 19 shows the effect of wind shear on σ_{zx} and Figure 20 shows the variation of σ_{zy}. It can be seen that the results obtained are very close to the results obtained by experiments. The values of σ_{zx} and σ_{zy} are very small because the vertical spread is small.

CHAPTER VI

DISCUSSION

The diffusion model developed in this thesis has wide application in practical air pollution studies. This model is capable of predicting concentration distribution not only in lower layers of the atmosphere but also at stratospheric heights. At lower altitudes the model is able to handle various stability conditions. Unlike other existing methods this model is able to give the time history of the diffusing cloud. All the methods which existed previously were able to give qualitative estimates at certain ranges of time and length scales. The various assumptions which have gone into the development of the diffusion equations used in the present model are based on experimental evidence. The final equations (2.119)-(2.124) are very simple to solve and do not require sophisticated numerical solution techniques. The set of equations give information about the cross correlation between different cloud coordinates which are nonzero when wind shear is present. This information was not available in the method developed by Kao (1962).

The present formulation requires information about the energy dissipation rate and the turbulent fluctuations. The number of measurements made for calculating these quantities are enormous but the atmospheric conditions are always changing and at an instant it is not possible to obtain the values of ϵ and σ's representative of the time. Thus it is always necessary to obtain an estimate of the above values. The

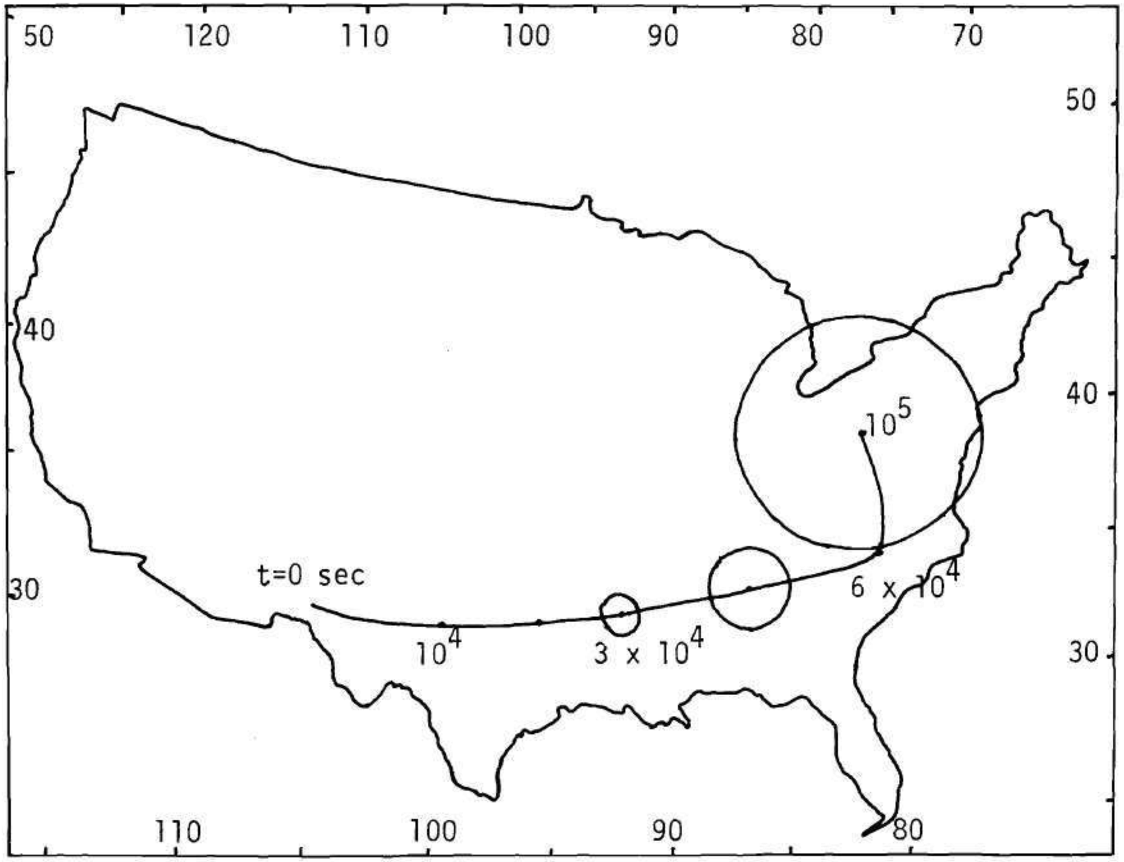


Figure 21. Diffusion During January at 32 KM Altitude.

TABLE 5: Time History of a Diffusing Cloud at 40 Km Altitude

Time $\times 10^{-4}$ sec	Latitude Deg	Longitude Deg	dU/dZ $\times 10^4$ sec^{-1}	dV/dZ $\times 10^4$ sec^{-1}	$\Gamma \times 10^4$ m^{-1}	$\epsilon \times 10^3$ m/sec^2	k sec^{-1}
0	32.50	106.5	3.4	1.1	4.98	3.	0.126
1	32.13	101.11	3.3	1.1	5.5	3.	0.126
2	32.75	96.31	3.1	0.6	5.5	3.	0.126
3	33.8	91.4	3.	0.6	5.3	3.	0.126
4	34.03	86.29	2.9	0.6	5.3	3.	0.126
5	33.95	83.35	2.9	0.6	5.4	3.	0.126
6	34.34	81.14	2.8	0.6	5.5	3.	0.126
7	35.21	79.52	2.7	0.6	5.5	3.	0.126
8	36.69	79.45	2.7	0.6	5.5	3.	0.126
9	38.55	79.93	2.6	0.6	6.1	3.	0.126
10	40.3	80.09	3.2	0.6	6.1	3.	0.126

Table 5 continued on next page.

TABLE 5: (continued)

Time $\times 10^{-4}$ sec	L_x km	L_z km	U m/sec	V m/sec	σ_u m/sec	σ_v m/sec	σ_x km	σ_y km	σ_z km
0	40.	4.4	64.	2.	24.	12.	.01	.01	.01
1	39.8	4.3	41.	3.	24.	12.	1.6	1.5	.1
2	39.9	4.4	33.	11.	24.	13.	33.4	30.6	1.1
3	39.9	4.4	63.	-2.	25.	13.	112.1	102.1	2.2
4	39.9	4.4	22.	-3.	25.	13.	194.2	176.9	2.9
5	39.9	4.4	7.	-4.	25.	13.	273.4	249.7	3.5
6	39.9	4.4	12.	8.	26.	13.	348.7	317.9	4.0
7	39.9	4.4	5.	8.	26.	14.	420.7	383.8	4.5
8	39.9	4.5	-23.	36.	27.	15.	490.9	448.1	4.9
9	39.9	4.5	0.	13.	29.	15.	560.1	511.9	5.4
10	39.8	4.6	-10.	9.	30.	16.	622.3	569.4	5.7

resulting solution of the diffusion equation will be accurate only within the accuracy of these estimates.

Another drawback in the present model is that it cannot be used to predict global diffusion, because of the fact that a locally horizontal mean motion was assumed in formulating the equations. Hence the solution can be applied regionally (over continental dimensions), but not around the globe.

An example of its application in the prediction of atmospheric air pollution is shown in figure 21. This shows the diffusion of a cloud which is released at Whitesands Missile Range during the month of January at an altitude of 40 kilometers. During this period of the year the general wind direction is from west to east and the cloud is carried eastward. Table 5 shows the time history of the diffusing cloud and the variations in the wind statistics. Most of the diffusion occurs only when it reaches the eastern coast. The GRAM-DIFFUSION program can be used to perform similar analyses during various times of the year and would be of immense value to air pollution meteorology.

Many of the results obtained for the specialized situations like small time behavior and large time behavior are all consistent with earlier derivations by numerous authors. The t^3 dependence for large time behavior was identical formally to those obtained by dimensional reasoning. The ϵt^3 dependence at small time was found to be negative in sign unlike the earlier belief of an additive contribution. Even though there is evidence for the existance of a positive t^3 variation it is either at intermediate or large times and is consistent with the present results, which indicate the significance of wind shear in producing t^3

diffusion at intermediate and large times.

The possibility of obtaining a parameter α for vertical diffusion needs further investigation. Once such a parameter is obtained the vertical diffusion prediction becomes very easy and will be of valuable help in the practical applications. The value of k as obtained in the present analysis could vary within an order of magnitude. But the present empirical results for cloud sizes are valid only within a factor of 2 and that accuracy is still maintained with this variation of k.

The present model has achieved its goal in being able to predict diffusion rates in the troposphere and stratosphere, within the accuracy prescribed by the accuracy of the available data. Even though it has not reached perfection it is hoped that enough foundation materials have been put together, for later developments.

APPENDIX

The two point second order correlation for velocities is important in the analysis of diffusion studies. It has been found that there are basically two different correlation functions in terms of which the entire array of two point correlations can be explained.

Longitudinal Correlation

As shown in Figures A1, this is a correlation between velocity components, which are aligned in the same direction as that the vector separation distance. Mathematically this can be explained in the following way

$$f(r) = \overline{U_p(\vec{x})\ U_p(\vec{x} + \vec{r})}/\overline{U_p^2} \qquad \text{(A-1)}$$

An interesting fact about the longitudinal correlation is that it is never negative. This is sometimes associated with the compressibility of the medium.

Lateral Correlation

Figure A-2 shows this type of correlation. Here the components of velocity are perpendicular to the vector separation distance. Mathematically this can be expressed as

$$g(r) = \overline{U_n(\vec{x})\ U_n(\vec{x} + \vec{r})}/\overline{U_n^2} \qquad \text{(A-2)}$$

the lateral correlation can be negative at large separation distances.

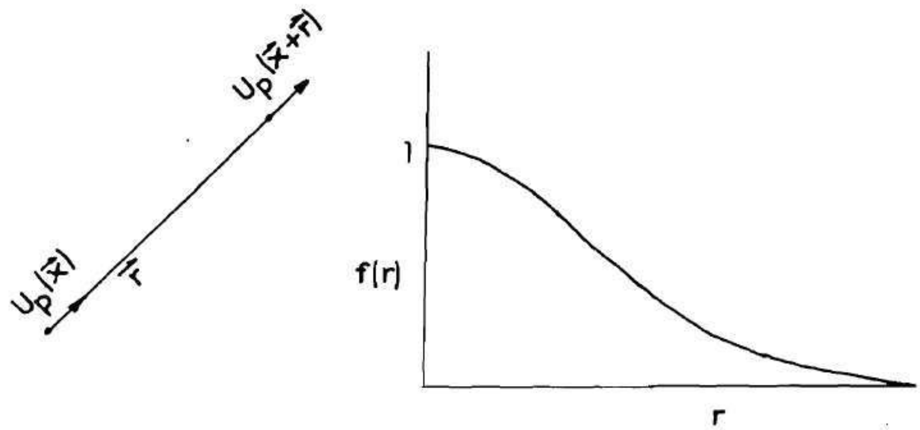


Figure 22. The Longitudinal Correlation.

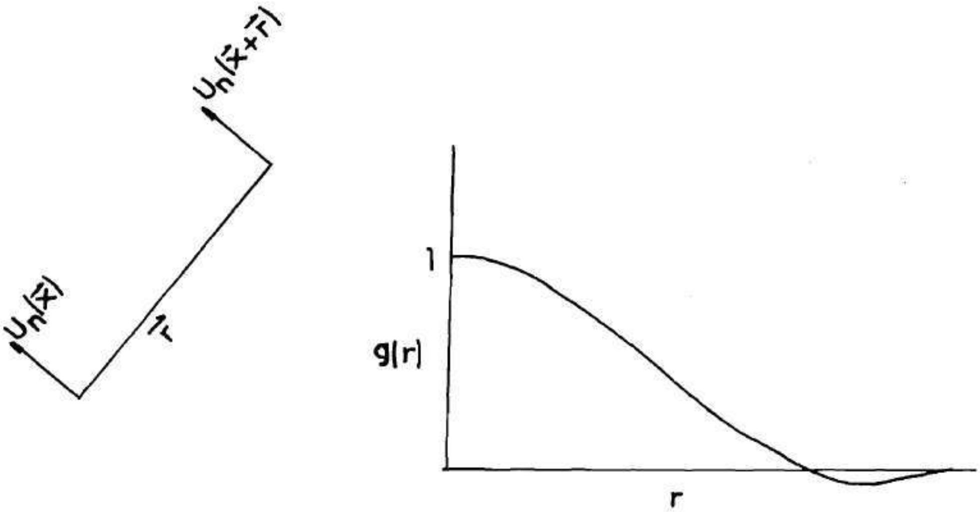


Figure 23. The Lateral Correlation.

The continuity equation for incompressible flows gives a relation between f(r) and g(r) in the following fashion.

$$g(r) = f(r) + \frac{1}{2}\, r f' \qquad \text{(A-3)}$$

The array of two point second order correlations can be then expressed in the following way.

$$R_{ij}(\vec{r}) = u^2\left(\frac{f - g}{r^2}\, r_i r_j + g\delta_{ij}\right) \qquad \text{(A-4)}$$

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